

**Laboratory Directed Research  
and Development Program  
FY 2001**

March 2002

## **Disclaimer**

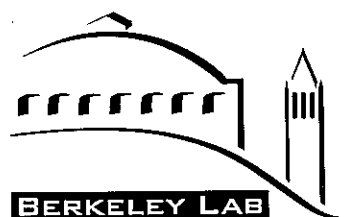
This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor The Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or The Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or The Regents of the University of California and shall not be used for advertising or product endorsement purposes.

Ernest Orlando Lawrence Berkeley National Laboratory  
is an equal opportunity employer.

**Report on  
Ernest Orlando Lawrence  
Berkeley National Laboratory**

# **Laboratory Directed Research and Development Program**

FY 2001



Ernest Orlando Lawrence  
Berkeley National Laboratory  
Berkeley, California 94720

**Office of Science  
U.S. Department of Energy**

This report is available on the World Wide Web at:  
[www.lbl.gov/Publications/LDRD/](http://www.lbl.gov/Publications/LDRD/)

Prepared for the U.S. Department of Energy under Contract No. DE-AC03-76SF00098



# Table of Contents

<b>Introduction .....</b>	<b>ix</b>
<b>Accelerator and Fusion Research Division.....</b>	<b>1</b>
Ian Brown Eleanor Blakely	Large Patterned Networks of Living Neurons ..... <b>1</b>
Phillip Colella Eric Esarey Alex Friedman Robert Ryne	Advanced Simulation of Complex Beam Systems..... <b>2</b>
John Corlett Kem Robinson Alexander Zholents	Feasibility Study of a Dedicated Permanent Magnet Synchrotron Light Source for Ultrafast X-Ray Science..... <b>3</b>
Wim Leemans Eric Esarey Alexander Zholents Max Zolotorev William Barletta	Attosecond Electron Bunches for Coherent Ionization and Excitation of Atoms ..... <b>6</b>
Ka-Ngo Leung Gordon Wozniak Rick Firestone Eleanor Blakely Steve Kahl Jasmina Vujic	Compact Deuterium-Deuterium (D-D) Neutron Generator for Moderator Design and Biological Research..... <b>7</b>
Robert Ryne	Large-Scale Simulation of High-Intensity Beams in Circular Accelerators..... <b>9</b>
Max Zolotorev Robert Schoenlein Alexander Zholents	A Source of Far-Infrared Radiation Based on a Material with an Optically Switched Permittivity ..... <b>10</b>
<b>Advanced Light Source Division .....</b>	<b>13</b>
M. Zahid Hasan Jonathan Denlinger James Underwood Zhi-xun Shen Zahid Hussain	Filling in a “Big Gap”: A Novel Momentum-Resolved Inelastic Emission Soft X-Ray Spectrometer for the Study of Strongly Correlated Quantum Systems ..... <b>13</b>
Michael Martin John Byrd Wayne McKinney David Robin Alexander Zholents Robert Schoenlein	Investigation of Coherent Far-Infrared Synchrotron Radiation at the Advanced Light Source ..... <b>14</b>

Howard Padmore Paul Alivisatos Raymond Jeanloz	High-Pressure Research at the Advanced Light Source.....	16
John Spence	Orbital Ordering in Colossal Magnetoresistance .....	17
<b>Chemical Sciences Division .....</b>		<b>19</b>
Ali Belkacem Thornton Glover Michael Prior	Laser-Assisted Photoabsorption of X-Rays by Atoms and Feasibility of a Femtosecond X-Ray Atomic Switch .....	19
Harvey Gould Hiroshi Nishimura	Development of a Neutral Molecule Synchrotron Storage Ring.....	20
Daniel Neumark	Photoionization Experiments on Atoms and Molecules Adsorbed onto Helium Droplets .....	21
Heino Nitsche Petra Panak	Study of Radionuclide-Bacterial Interaction Mechanisms .....	21
<b>Computing Sciences (Information and Computing Sciences Division, National Energy Research Scientific Computing Division, and Mathematics Department) .....</b>		<b>25</b>
David H. Bailey Xiaoye Li	High-Precision Arithmetic and Applications .....	25
Grigory Barenblatt	Nonlinear Mathematical Models of Phenomena Related to Petroleum, Mining, and Geological Engineering.....	26
John Bell Phillip Colella Alexandre Chorin Nancy Brown	Science-Based Subgrid Scale Modeling in Fluid Turbulence.....	27
E. Wes Bethel S. Jacob Bastacky	Interactive Stereo Electron Microscopy Enhanced with Virtual Reality.....	29
Andrew Canning Steven Louie	Computational Methods for Electronic Structure Codes in Materials Science: Ground-State and Excited-State Properties.....	31
Niels Jensen	Atomic-Scale Modeling of Materials Chemistry.....	32
Xiaoye Li Leonid Oliker Katherine Yelick	Suitability of Alternative Architectures for Scientific Computing in the NERSC-5 Time Frame.....	33
Paul Hargrove William Saphir Robert Lucas	Berkeley Lab Distribution (BLD): Software for Scalable Linux Clusters .....	34
C. William McCurdy Thomas Rescigno	Electron Collision Processes above the Ionization Threshold .....	35

Gregory Miller	Second-Order Methods for Solid-Fluid Shock Coupling with Application to Martian Meteorites .....	37
Ekow Otoo	Adaptive File Replication and Coordinated Transfer for Data-Intensive Grid Applications.....	39
Bahram Parvin Qing Yang	Feature-Based Knowledge Discovery from Multidimensional Data Set .....	40
Horst Simon	Linear Algebra and Statistical Algorithms for Text Classification in Large Databases.....	42
<b>Earth Sciences Division .....</b>		<b>45</b>
James Bishop	Computer Tools and Methods for Demonstrating a Robotic Ocean Carbon Observing System.....	45
Inez Fung Horst Simon Jim Bishop Don DePaolo	Carbon-Water-Climate Interactions.....	45
Terry Hazen Curtis Oldenburg Sharon Borglin Peter Zawislanski	Aerobic Bioremediation of Landfills .....	48
Larry Myer Sally Benson	Geological Sequestration of Carbon Dioxide .....	50
Margaret Torn	Isotopic Analysis of Carbon Cycling and Sequestration in a California Grassland under Global Change Conditions.....	52
<b>Engineering Division .....</b>		<b>55</b>
Henry Benner Ronald Krauss	Fast Lipoprotein Identification Process .....	55
<b>Environmental Energy Technologies Division .....</b>		<b>57</b>
Hashem Akbari Paul Berdahl	Development of Cool Colored Shingles .....	57
Ronald Cohen Nancy Brown Allen Goldstein Robert Harley Haider Taha	Atmospheric Chemistry: Changes at the Interface between Urban and Regional Scales .....	58
William Fisk Regine Goth-Goldstein	Indoor Bioaerosol Detection and Quantification by Polymerase Chain Reaction (PCR).....	60
Donald Lucas Arlon Hunt	Diesel Particle Detection and Control .....	61

<b>Life Sciences Division</b> .....	63
Mary Helen Barcellos-Hoff Damir Sudar Bahram Parvin Abby Dernburg Daniel Callahan Richard Schwartz	Quantitative Spatial and Temporal Resolution of Multicellular Interactions... 63
Anat Biegon	Positron Emission Tomography (PET) Ligands for the N-methyl-D-aspartate (NMDA) Receptor Channel ..... 65
Mark Biggin	A High Sensitivity <i>In-Vivo</i> Crosslinking Method ..... 66
David Chen	Transgenic Mouse Models for DNA Damage Sensing, Repair, and Aging ..... 67
Abby Dernburg	Dynamic Reorganization of Chromosome Architecture during Meiosis..... 69
Michael Eisen	Experimental and Computational Analysis of the Mechanisms and Logic of Eukaryotic Transcriptional Regulation ..... 70
Carolyn Larabell	Tracking Proteins in Light and Soft X-Ray Microscopy..... 72
<b>Materials Sciences Division</b> .....	75
Carolyn Bertozzi	Functional Bone-Like Materials: A Biomimetic Synthetic Approach..... 75
Daniel Chemla	Condensation of Indirect Excitons in Coupled Quantum Well Nanostructures ..... 76
Michael Crommie	Nanoscale Transport in Ultra-Thin Films ..... 77
J.C. Séamus Davis	Atomically Resolved Spin-Polarized Imaging with Superconducting STM Tips ..... 79
Oscar Dubón, Jr.	Surfactant-Mediated Epitaxy of IV-IV Compounds: Expanding the Limits of Alloy Composition and Nanostructure Synthesis with Lead Overlayers..... 80
Charles Fadley Malcolm Howells	Holographic Imaging with X-Rays: Fluorescence and Fourier Transform Holography ..... 81
Paul Alivisatos Jean Fréchet	A Molecular Foundry: Complex Nanosystems for Designed Structure and Function ..... 83
Miquel Salmeron Frank Olgetree Charles Fadley Zahid Hussain	High Pressure Photoelectron Spectroscopy for Catalysis, Semiconductor Processing, and Environmental- and Bio-Chemistry..... 84
Yuen-Ron Shen	Laser Spectroscopy on New Materials: Theory and Experiment ..... 87
Shimon Weiss	Single-Molecule Protein Dynamics ..... 87
Peidong Yang	Infinite [Mo <sub>3</sub> Se <sub>3</sub> ] Chains as Molecular Conductors and Their Assemblies..... 89



<b>Nuclear Science Division .....</b>	<b>93</b>
I-Yang Lee Kai Vetter	Design of Digital Signal Processing Electronics for High-Resolution Solid-State Detectors..... 93
Heino Nitsche Uwe Kirbach	First Chemical Study of Element 108, Hassium ..... 94
<b>Physical Biosciences Division .....</b>	<b>97</b>
Steven Brenner Stephen Holbrook	SCOR: A Structural/Functional Classification of RNA..... 97
David Chandler	Development and Application of the General Theory of Hydrophobicity to Interpret Stability and Dynamics of Biological Assemblies ..... 99
Thomas Earnest	Molecular Recognition and Protein/Protein Interactions in Signal Transduction ..... 100
Robert Glaeser Kenneth Downing Eva Nogales Esmond Ng Ravi Malladi	Teraflop Challenges in Single-Particle Electron Crystallography ..... 101
Jay Groves Carolyn Bertozzi Arup Chakraborty	Cooperative Effects Determining Fidelity in Cellular Recognition ..... 102
Teresa Head-Gordon	Novel Synchrotron Experiments to Determine Hydration Forces for Molten Globules and Model Proteins for Extremophiles..... 104
Ehud Isacoff	Genetically Encoded Optical Sensors of Cell Signaling..... 105
<b>Physics Division .....</b>	<b>109</b>
Ian Hinchliffe Murdock Gilchriese	Modeling of High Energy Physics Detectors..... 109
Adrian Lee Helmuth Spieler	POLARBEAR: An Experiment to Measure Polarization Anisotropy in the Cosmic Microwave Background..... 109
Michael Levi Saul Perlmutter	Foundations for a SuperNova/Acceleration Probe (SNAP)..... 111
	Sub-Project 00026: Development of an Advanced Imaging Array..... 111
	Sub-Project 00027: Nearby Supernova Search with Ten-Fold Increase in Efficiency ..... 112
George Smoot Jodi Lamoureux	Large Astrophysical Data Sets..... 114
<b>Acronyms and Abbreviations .....</b>	<b>117</b>



# Introduction

The Ernest Orlando Lawrence Berkeley National Laboratory (Berkeley Lab or LBNL) is a multi-program national research facility operated by the University of California for the Department of Energy (DOE). As an integral element of DOE's National Laboratory System, Berkeley Lab supports DOE's missions in fundamental science, energy resources, and environmental quality. Berkeley Lab programs advance four distinct goals for DOE and the nation:

- To perform leading multidisciplinary research in the computing sciences, physical sciences, energy sciences, biosciences, and general sciences in a manner that ensures employee and public safety and protection of the environment.
- To develop and operate unique national experimental facilities for qualified investigators.
- To educate and train future generations of scientists and engineers to promote national science and education goals.
- To transfer knowledge and technological innovations and to foster productive relationships among Berkeley Lab's research programs, universities, and industry in order to promote national economic competitiveness.

Berkeley Lab's research, in general, and the LDRD program, specifically, all unclassified, support DOE's important science mission, as articulated in the Secretary of Energy's FY 2003 Budget Rollout. This statement includes that "we...want to use the talents nurtured by our science program to leapfrog today's energy security problems," and that "as the irreplaceable foundation for tomorrow's security demands, we need a strong physical science program." Furthermore, "DOE's program in science is a story of immense accomplishments and vast promise...We are leaders in pursuit of the basic understanding of matter and physical science." In addressing fundamental research, the statement further notes that "our work to better understand the fundamentals of energy production and its impacts has had far-reaching applications in biology, chemistry, nuclear medicine, and materials science." The LDRD program supports these goals; and some of the research referenced in the Secretary's statement has its origins in the LDRD program.

The *Berkeley Lab Laboratory Directed Research and Development Program FY 2001* report is compiled from annual reports submitted by principal investigators following the close of the fiscal year. This report describes

the supported projects and summarizes their accomplishments. It constitutes a part of the Laboratory Directed Research and Development (LDRD) program planning and documentation process that includes an annual planning cycle, project selection, implementation, and review.

The Berkeley Lab LDRD program is a critical tool for directing the Laboratory's forefront scientific research capabilities toward vital, excellent, and emerging scientific challenges. The program provides the resources for Berkeley Lab scientists to make rapid and significant contributions to critical national science and technology problems. The LDRD program also advances Berkeley Lab's core competencies, foundations, and scientific capability, and permits exploration of exciting new opportunities. All projects are work in forefront areas of science and technology. Areas eligible for support include the following:

- Advanced study of hypotheses, concepts, or innovative approaches to scientific or technical problems;
- Experiments and analyses directed toward "proof of principle" or early determination of the utility of new scientific ideas, technical concepts, or devices; and
- Conception and preliminary technical analyses of experimental facilities or devices.

The LDRD program supports Berkeley Lab's mission in many ways. First, because LDRD funds can be allocated within a relatively short time frame, Berkeley Lab researchers can support the mission of the Department of Energy (DOE) and serve the needs of the nation by quickly responding to forefront scientific problems. Second, LDRD enables Berkeley Lab to attract and retain highly qualified scientists and supports their efforts to carry out world-leading research. In addition, the LDRD program also supports new projects that involve graduate students and postdoctoral fellows, thus contributing to the education mission of Berkeley Lab.

Berkeley Lab has a formal process for allocating funds for the LDRD program. The process relies on individual scientific investigators and the scientific leadership of Berkeley Lab to identify opportunities that will contribute to scientific and institutional goals. The process is also designed to maintain compliance with DOE Orders, in particular DOE Orders 413.2 dated March 5, 1997, and 413.2A, dated January 8, 2001. From year to year, the distribution of funds among the scientific program areas

changes. This flexibility optimizes Berkeley Lab's ability to respond to opportunities.

Berkeley Lab LDRD policy and program decisions are the responsibility of the Laboratory Director. The Director has assigned general programmatic oversight responsibility to the Deputy Director for Research. Administration and reporting on the LDRD program is supported by the Directorate's Office for Planning and Strategic Development. LDRD accounting procedures and financial management are consistent with the Laboratory's accounting principles and stipulations under the contract between the University of California and the Department of Energy, with accounting maintained through the Laboratory's Chief Financial Officer.

In Fiscal Year (FY) 2001, Berkeley Lab was authorized by DOE to establish a funding ceiling for the LDRD program of \$14.2 M, which equates to about 3.5% of Berkeley Lab's FY01 projected operating and capital equipment budgets. This funding level was provided to develop new scientific ideas and opportunities and allow the Berkeley Lab Director an opportunity to initiate new directions. Budget constraints limited available resources, however, so only \$9.6 M was expended for operating and \$0.6 M for capital equipment (2.7% of actual FY01 costs).

In FY01, scientists submitted 135 proposals, requesting over \$23.9 M in operating funding. Sixty-nine projects were funded, with awards ranging from \$25 K to \$600 K. These projects are summarized in Table 1.

# Accelerator and Fusion Research Division

---

## Large Patterned Networks of Living Neurons

---

Principal Investigators: Ian Brown and Eleanor Blakely

Project No.: 01001

### Project Description

While techniques such as magnetic resonance imaging and positron emission tomography have provided insight into the location of neural activity within the brain, the minimum resolvable volume (~1 cubic millimeter) contains  $\sim 10^3$  to  $10^4$  neurons. In order to learn the details of how large systems of neurons communicate, we first need to develop methods for growing networks of large numbers of live neurons whose dendrite and axon connections can be controlled in predetermined ways, and second to develop means for stimulating and monitoring excitation. Success with this challenge will be of great importance to our understanding of the working of the human brain and peripheral nervous system, and to novel kinds of computer architecture. Large *in-vitro* networks could show, for example, the emergence of stable patterns of activity, and could lead to an understanding of how groups of neurons learn after repeated stimulation.

Our approach to patterning will be to use selective ion implantation or plasma deposition on the substrate on which the neural array will subsequently be grown, using ion species that enhance or inhibit neural growth. We will explore growth-stimulating and growth-inhibiting surface modification methods so as to learn the most effective method (highest "contrast"). Lithographic masks of various feature sizes and network patterns will be used to form the arrays. Our approach to neuron excitation will be to use extra-cellular electrical stimulation through electrically conducting, optically transparent, plasma deposited ultra-thin metal films. We will also explore the simultaneous recording of self-generated action-potentials of large numbers of neurons throughout the network by novel embodiments of charge-coupled device (CCD) detector arrays used for extra-cellular voltage pick-up.

### Accomplishments

Methods for selectively enhancing or inhibiting neuron growth, both by energetic ion implantation and by plasma

deposition, were the primary object of the work for the first year of the project. A basic and fundamentally important primary result was that there are indeed great differences between the effects due to different ion species—this approach is thus in principle a quite valid approach for altering the neuro-compatibility of the substrate on which the neural culture is to be grown.

Neuron cultures were grown, monitored, and quantified on substrates that had been ion implanted or plasma deposited using a range of plasma/ion beam parameters, particularly variation of the ion species. We determined that the best plasma processing, in terms of enhancing selective neuron growth, was the formation of a super-thin (of order 100 angstrom) layer of diamond-like carbon atomically mixed into the substrate surface. Some test patterns (with rather large feature-size in this preliminary work) of diamond-like carbon were made, and we were able to subsequently grow neurons (PC-12 rat neurons) on the pattern. The neuron growth "patterning contrast ratio" (ratio of neuron density on the plasma-treated regions to neuron density on the untreated regions) was very high, with prolific growth on the plasma-treated regions and virtually none on the untreated regions. Good contrast was also observed for tantalum ion implantation. We hope to pursue this part of the work for its fundamental importance to cell attachment biology as we continue with the main program. For our present purposes, we have selected diamond-like carbon as our preferred neural-growth-enhancing substrate pre-treatment.

We also started on our exploration of the use of CCD chips as substrates onto which the patterned neural arrays are grown. Our hope here is that the individual CCD detector elements can provide a pick-up mechanism for detecting action potential activity in the neurons growing on them. This novel approach has the immense advantage of making use of existing technology to provide the large array of pick-up electrodes, associated electronics, and display of the CCD-derived space-time pattern of neural signal activity. To accomplish this, the protective glass layer (~0.5 millimeter thick) that covers the CCD elements themselves needs to be removed, and replaced by a super-thin (<1000 angstroms) neuro-compatible plastic film. Techniques for managing this high-precision work are being developed.

### Publications

E.A. Blakely, K.A. Bjornstad, J.E. Galvin, O.R. Monteiro, and I.G. Brown, "Selective Neuron Growth on Ion Implanted and Plasma Deposited Surfaces," abstract submitted to *IEEE 29<sup>th</sup> International Conference on Plasma Sciences*, Banff, Canada, (May 26-30, 2002).

---

## **Advanced Simulation of Complex Beam Systems**

---

Principal Investigators: Phillip Colella, Eric Esarey, Alex Friedman, and Robert Ryne

Project No.: 01002

### **Project Description**

This work has two important strategic goals:

- First, to provide a computational foundation for the two key scientific areas critical to the development of the next generation of particle and laser-beam based accelerators: (1) wave-particle interaction in the full six-dimensional phase-space of multi-particle, multi-mode particle-wave systems; and (2) coulomb and electromagnetic space-charge-dominated transport of high-intensity, charged particle beams. These will have immediate applications in the studies of laser-plasma acceleration, acceleration and transport of heavy ions for fusion, and simulations of intense bunches in circular accelerators including three-dimensional (3D) space-charge effects.
- Second, to build a capability that combines accelerator physics with applied mathematics, numerical algorithms, and advanced computing, to address the nation's next-generation programs in accelerator science and technology.

This project brings together theoretical and experimental beam physicists, computational scientists, and computer scientists to address some of the most critical problems in beam science. Although all the accelerator/beam systems have unique features that require special techniques for simulation, there are many computational tools and techniques that are common to all these systems and to many others. Our approach is to identify and develop tools that can be adopted to meet the needs of a broad spectrum of particle beam simulation projects. The primary focus will be on tools that significantly enhance code performance, permit codes to be readily ported to the highest performance machines at the National Energy Research Scientific Computing Center (NERSC) and elsewhere, and allow accelerator scientists to use advanced visualization techniques to understand complex beam systems. These

tools will be turned into components that can be used by a number of simulation projects. Some of the techniques to be developed are general, and will have applicability to a broad range of emerging research areas at Berkeley Lab and elsewhere.

Particle-in-cell simulations of laser and plasma-based advanced accelerators will be carried out based on the code XOOPIC. Originally, XOOPIC was developed for a variety of non-accelerator applications (e.g., plasma processing, plasma display panels, microwave tubes). Under this program, XOOPIC will be modified to allow efficient simulation of advanced accelerators. This will include implementation of a moving window, i.e., a simulation domain that moves with the beam at the speed of light; generalization of the electron beam and laser pulse launchers; inclusion of collisional models valid for highly relativistic electrons; and further optimization for efficient performance on massively parallel platforms. Simulation of relevant advanced accelerator configurations will be carried out.

We will complete the integration of the WARP particle-in-cell beam simulation code with the Adaptive Mesh Refinement (AMR) package Chombo, via a scripting level interface based on the Python language. This requires significant modification of both code packages, and development of the data-passing and parallel domain-decomposition management. Introducing AMR into codes for accelerator modeling has the potential for reducing by an order-of-magnitude or more the amount of computer time required to perform such calculations. Using the new capability, we will carry out simulations of space-charge dominated beams in injectors and accelerating structures with greater fidelity and efficiency than has heretofore been possible, with relevance to a number of applications for advanced accelerators.

### **Accomplishments**

#### *Simulation of Plasma-Based Advanced Accelerators:*

The code XOOPIC has been modified to enable particle-in-cell simulations of plasma-based accelerators and plasma lenses, including the effects of relativistic electron-neutral collisions. These modifications include the development of a moving window algorithm, adding a new electromagnetic pulse launcher, generalization of the particle beam emitters, and further optimization to allow efficient use on parallel platforms. Also, new relativistic parametric models for impact ionization and elastic scattering have been developed and implemented for lithium and nitrogen. These include total cross section, energy distribution of secondary electrons, and scattering angles models.

For example, the original version of XOOPIC utilized a static (stationary in the lab frame) simulation domain. For

accelerator applications, it is necessary to utilize a moving window, i.e., a simulation domain that moves at the speed of light. In this case, the simulation domain follows the beam, and only particles in the region near the beam are retained in the simulation, thereby greatly decreasing the memory and computation time required. To demonstrate the utility of XOOPIC, we performed two-dimensional simulations of both laser driven [the laser wake field accelerator (LWFA)] and electron beam driven [the plasma wake field accelerator (PWFA)] plasma-based accelerators. For the LWFA, simulations were carried out in two-dimensional slab geometry, for both low- and high-intensity laser pulses. Excellent agreement was obtained with both theoretical models (low-intensity case, for which analytical solutions exist) and with nonlinear fluid simulations (high-intensity case). These simulations have relevance to the experiments being carried out by the I'OASIS (laser Optics and Acceleration Systems Integrated Studies) Group of Berkeley Lab.

Simulations of the PWFA were carried out in two-dimensional cylindrical geometry with parameters relevant to the E-157 experiments at Stanford Linear Accelerator Center (SLAC). In the E-157 experiments, the 30 GeV SLAC electron beam is propagated through a 1 meter long lithium plasma (partially ionized), generating wakefields with accelerating fields on the order of 1 gigavolt per meter. Simulations with XOOPIC confirm the generation of large wakefields, and were in good agreement to results obtained with the particle-in-cell (PIC) code OSIRIS used by the University of California, Los Angeles group. In addition, effects of electron-neutral collisions on the PWFA were studied using the new lithium collisional models implemented in XOOPIC. These simulations indicate that the additional beam-induced ionization of lithium was not significant for the present E-157 experiments. However, for future PWFA experiments at SLAC on the so-called "plasma afterburner" concept, which utilizes a 100-times higher lithium density, simulations indicate that the additional beam-induced ionization can greatly reduce the expected wake amplitude. These and other effects are currently under study.

#### *Space-Charge-Dominated Transport of High-Intensity Charged Particle Beams*

A nodal implementation of a multigrid Adaptive Mesh Refinement (AMR) solver for the Poisson equation using Shortly-Weller ("cut cell") discretization of the Laplacian operator, to account for internal boundaries at subgrid resolution, was developed, and an initial coupling to the PIC accelerator beam dynamics code WARP3D was implemented. Using analysis and simulations, we studied the interaction of a simulation macroparticle with a locally refined mesh. We confirmed our expectation that the anisotropy of the grid topology associated with local mesh refinement results in anomalous self-forces on macroparticles and makes linearly varying fields become

non-linear (anharmonic). The first effect is of importance for any plasma code using AMR, while the second is of particular significance for accelerator codes where preservation of the beam quality (which is guaranteed when forces are linear in the spatial coordinates) is a key issue. Our goal is to ensure that the spurious forces that arise do not lead to an unacceptable level of inaccuracy. We learned that, by careful choice of the method by which fields are interpolated onto the particles, it is possible to greatly reduce the size of such anomalous forces.

We studied the sensitivity of the beam in a simulated ion diode to the details of the boundary conditions applied on the outer edge of a subdomain surrounding the beam, and established the need for AMR in this application. Thus, we expect that PIC simulations using AMR will indeed achieve the improvements in accuracy and efficiency that were anticipated at the initiation of this project.

#### **Publications**

D.L. Bruhwiler, R.E. Giacone, J.R. Cary, J.P. Verboncoeur, P. Mardahl, E. Esarey, W. Leemans, and B.A. Shadwick, "Particle-in-Cell Simulations of Plasma Accelerators and Electron-Neutral Collisions," *Physical Review Special Topics, Accelerators and Beams*, **4**, 101302 (2001) <http://prst-ab.aps.org/abstract/PRSTAB/v4/i10/e101302>.

D.L. Bruhwiler, R.E. Giacone, J.R. Cary, J.P. Verboncoeur, P. Mardahl, E. Esarey, and W. Leemans, "Modeling Beam-Driven and Laser-Driven Plasma Wakefield Accelerators with XOOPIC," *Advanced Accelerator Concepts*, **569**, 591-604 (AIP 2001).

---

### ***Feasibility Study of a Dedicated Permanent Magnet Synchrotron Light Source for Ultrafast X-Ray Science***

---

Principal Investigators: John Corlett, Kem Robinson, and Alexander Zholents

Project No.: 01003

#### **Project Description**

We have identified a promising technique for generation of femtosecond x-ray pulses of synchrotron radiation. Development of this technique may result in the design and construction of a new dedicated synchrotron light source where ultrafast x-ray science can be conducted at a large

number of the beamlines with the average flux of the femtosecond x-ray pulses of the order of  $10^{10}$  photons/second. Our original proposal involved the generation of femtosecond x-ray pulses from beams circulating in a storage ring —this scheme has evolved during the initial stages of our study and now is optimized in a recirculating linac design.

The goal demands x-ray pulses with ultra-short time duration, and for typical experiments a large flux is more important than high brightness. Our proposed source has the short pulse length necessary ( $\sim 60$  femtoseconds FWHM) to study very fast dynamics, high flux to study weakly scattering systems, and tuneability over 1 to 10 keV photon energy. The repetition rate is determined by either the time required for the experimental sample to return to its ground state (often limited by acoustic propagation velocities), the time to translate the sample to a new area after each shot, or the repetition rate of the attendant pump laser systems.

Our studies have resulted in a proposed facility that can be built on a basis of existing technologies, to produce femtosecond x-ray pulses with high flux, and repetition rate matched to the requirements of structural dynamics experiments. The facility uses a recirculating linac for acceleration (and deceleration) of electrons produced by a high-brightness photocathode radiofrequency (rf) gun, at a bunch repetition rate of approximately 10 kHz. The system design produces a small vertical emittance, which is key to producing the  $\sim 60$  femtosecond x-ray pulses. After acceleration through a superconducting recirculating accelerator, the electron pulses are compressed in the final arc to  $\sim 1$  picosecond (ps) duration. A specialized technique is then employed to produce the femtosecond x-ray pulses by first imprinting on the electron bunches a transverse momentum that is correlated to longitudinal position within the bunch. This action results in an emitted x-ray pulse with a time-correlated spatial and angular distribution. The correlated x-ray pulse is then compressed by use of asymmetrical crystal optics to achieve the ultimate photon pulse length.

## Accomplishments

A layout of the present machine concept is shown in the Figure. It consists of an rf photo-injector, a linear pre-accelerator, a main linear accelerator, magnetic arcs and straight sections, and a photon beam production section. Electron pulses of  $\sim 10$ -picosecond duration are produced in a high-brightness rf photocathode gun and accelerated to 10 MeV. Application of a solenoidal magnetic field in the cathode region, followed by a skew-quadrupole channel, allows production of a "flat" beam with x/y emittance ratio  $> 50$ . The electrons are further accelerated in a 1.3 GHz superconducting linac pre-accelerator to 120 MeV, and then injected into a recirculating linear accelerator. Passing

the arc connecting the pre-accelerator and the recirculating linac the electron bunches are compressed to approximately 5-picosecond full length. In the recirculating linac the final energy of  $\sim 2.5$  GeV is achieved after four passes through the 600 MeV superconducting linac. The final arc compresses bunches to an approximately 1-picosecond length, and the bunches receive a time-correlated head-tail vertical kick in a dipole-mode rf cavity. The electrons then radiate x-rays in undulators and dipole magnets. There are seven two-meter long straight sections for undulators as well as six 2-tesla field dipole magnets located between undulators. After passing through the photon production section, the electrons receive a compensating time-correlated head-tail vertical kick, and are stretched back to approximately 5 picoseconds. They are transported back to and decelerated in the recirculating accelerator structure and the 110 MeV pre-accelerator to  $< 10$  MeV and turned into a beam dump. This deceleration reduces the design challenges and radiation hazard of the beam dump. Though the electron beam does return almost all of its energy to the linac rf fields, the circulating beam power is so low that it does not contribute significantly to the overall system efficiency.

A "beam spreader" section separates the beam at various energies into their respective transport paths. Due to the proximity of beamlines of different energies, specialized magnet designs and careful layout of components are employed in this section. Apart from specialized magnets in the beam spreader section, the magnets are of conventional electromagnet design similar to those of existing synchrotron radiation sources.

At the beginning and end of the photon production section, we use superconducting rf cavities operating in the first dipole mode ( $TM_{110}$ ) to prepare the electron beam to allow the x-ray compression from  $\sim 1$  picosecond to  $\sim 60$  femtoseconds. The first cavity provides a time dependent transverse (vertical) kick to the electron bunches that the second cavity cancels. The rf phase is adjusted so that the kick acts in opposite directions on the head and the tail of the bunches, with no perturbation to the center of the bunch. The electrons perform betatron oscillations after the kick with vertical betatron phase advance of  $n\pi$  between the center of the rf cavity and the center of the  $n$ -th undulator. The vertical betatron phase advance is  $n\pi/2$  between the center of the undulator and the center of the adjacent 2-tesla dipole magnets. Therefore within a bending magnet the electrons in a bunch have zero angle and a transverse displacement variation along the bunch. Similarly, we find in the insertion devices that the electrons within a bunch have no transverse displacement and an angular variation along the bunch. Consequently, the photon production bend magnets are x-ray sources with a vertical position-time correlation and the insertion devices are x-ray sources with an angular-time correlation, and these correlations are exploited to compress the x-ray pulses. The design



maintains the ability to vary lattice functions over a wide range to control the position and angular correlation.

Asymmetrically cut crystals may be used as optical elements in the x-ray pulse compression scheme. As a result of the different angles of incidence and diffraction, a crystal may be oriented to produce a variable path length across the x-ray beam, and two asymmetrically cut crystals can be used as both monochromator and pulse compressor for the x-rays.

Emittance control and understanding and mitigation of collective effects is critical to a successful machine design, and we are addressing key aspects of accelerator physics involved in beam break-up, coherent synchrotron radiation, the influence of resistive wall wakefields, and other effects.

Our approach for producing a short x-ray pulse relies on the availability of an electron beam source with a flat aspect ratio so that one transverse dimension (y, vertical) is small. This principal has been successfully demonstrated at the Fermilab/ Northern Illinois Center for Accelerator and

Detector Development (NICADD) Photoinjector Laboratory (FNPL). The successful demonstration at the FNPL of flat beams with emittance ratio 50:1 is highly encouraging and we have joined the experiment collaboration to help further work in this regard.

Conventionally, photocathode rf guns employ a half-length pillbox cell for the cathode cavity followed by a full cell for rapid acceleration of emitted electrons. For continuous wave (CW) or high duty factor operation, thermal limitations may prevent such a design from operating at sufficiently high gradient. We have produced a preconceptual design with optimized cavity geometry to allow cooling of the cavity surfaces, and operation at high gradient and high repetition rate. In this design the first cell is modified by the inclusion of a re-entrant nosecone, on the end face of which the photo-cathode is mounted. This nosecone serves two purposes: it increases surface area to reduce deposited power density, and it enhances the accelerating electric field at the cathode.

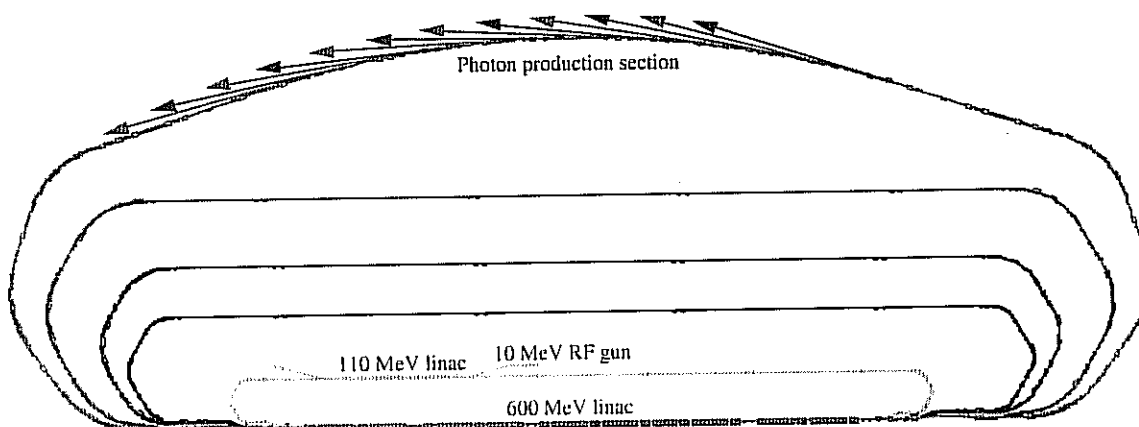


Figure 1: Recirculating linac for femtosecond x-ray production.

## Publications

J. N. Corlett, S. DeSantis, N. Hartman, P. Heimann, R. LaFever, D. Li, H. Padmore, R. Rimmer, K. Robinson, R. Schoenlein, J. Tanabe, S. Wang, and A. Zholents, "Initial Feasibility Study of a Dedicated Synchrotron Radiation Light Source for Ultrafast X-Ray Science," LBNL 48171, (October 2001) <http://www-library.lbl.gov/docs/LBNL/48171/PDF/LBNL-48171.pdf>

J. N. Corlett, S. DeSantis, N. Hartman, P. Heimann, R. LaFever, D. Li, H. Padmore, R. Rimmer, K. Robinson, R. Schoenlein, J. Tanabe, S. Wang, A. Zholents, and D. Kairan, "A Dedicated Synchrotron Light Source for Ultrafast X-Ray Science," to be published in *Proceedings of 2001 Particle Accelerator Conference*, Chicago, Illinois, (June 2001) <http://www-library.lbl.gov/docs/PUB/5466/PDF/PUB-5466.pdf>

---

## Attosecond Electron Bunches for Coherent Ionization and Excitation of Atoms

---

Principal Investigators: Wim Leemans, Eric Esarey, Alexander Zholents, Max Zolotarev, and William Barletta

Project No.: 01004

### Project Description

Use of attosecond electron bunches offers the exciting possibility of new insights into our understanding of atomic dynamics such as wave function collapse and decoherence of quantum states. An attosecond bunch has an electric field, which is a truly sub-cycle (as opposed to multi-cycle radiation pulses), unipolar pulse with a broad spectrum, due to its shortness, containing photons with energy that can be comparable to the transition energy of atomic electrons or even ionization energy. Coherent excitation and ionization of atoms by these collective fields is expected to occur in interaction with media. Coherent ionization losses are expected to significantly reduce stopping ranges for relativistic electrons and, e.g. can be used for measurements of the duration of attosecond bunches. This project will attempt to address the following: (1) methods for production of attosecond bunches, and (2) development of a theoretical understanding of coherent ionization and coherent excitation processes and their potential applications. Laser-plasma acceleration, direct ponderomotive acceleration, and laser-electron beam slicing will all be considered as potential sources.

Tools will be developed to design, model, and analyze sources capable of producing attosecond electron bunches. Experiments will be initiated utilizing femtosecond bunches produced via laser-plasma interaction. Efforts are underway to demonstrate production of 1 to 3 femtosecond bunches using the colliding laser pulse method. Three-dimensional test particle simulations show production of ultrashort (1 femtosecond), energetic (10s of MeVs), low energy spread (0.3%) bunches by using three high-power (1 to 10 terrawatt) ultrashort (40 to 50 femtoseconds) laser pulses in a 1-millimeter plasma length. Preliminary analysis of the ponderomotive vacuum acceleration indicates a true potential for production of attosecond bunches. Bunch lengthening due to finite energy spread and space charge effects will be evaluated. Laser-electron beam slicing was recently demonstrated at the Advanced Light Source and its potential for production of attosecond bunches will also be evaluated.

### Accomplishments

We have carried out theoretical studies of three methods for generating femtosecond or attosecond electron bunches. The first method relied on the use of a laser wakefield accelerator (LWFA) operating in the standard regime with a typical density on the order of  $n_e = 10^{18} \text{ cm}^{-3}$  and a plasma wavelength on the order of 30 microns (100 femtoseconds). For a mono-energetic electron bunch, injected into a wakefield, to be accelerated while maintaining a small energy spread, it is necessary that the bunch occupies a small fraction of the wake period, i.e., the bunch must be ultra-short, on the order of a few femtoseconds. To analyze the injection of such bunches with the colliding pulse method, in which two counterpropagating laser pulses are used to inject electrons from the background plasma directly into the wake, two different approaches have been taken. The first relied on test particle simulations of the colliding pulse LWFA injector using prescribed fields. These simulations indicated the production of a trapped bunch containing  $10^7$  electrons with a low energy spread ( $< 1\%$ ), low normalized emittance ( $< 1 \text{ mm-mrad}$ ), and of ultra-short duration (order of 1 femtosecond). Such test particle simulations, however, used idealized prescribed fields and neglected the space charge effects of the accelerated bunch.

To address the first shortcoming, particle-in-cell code simulations were carried out using the two-dimensional fully relativistic code XOOPIC (Object Orient Particle in Cell code for X11-based Unix workstations). These simulations were carried out by researchers at the University of Colorado, Boulder, in collaboration with researchers here at Berkeley Lab (Leemans, Esarey *et al.*). The code was upgraded to allow injection of multiple laser beams (co- and counter propagating). In addition, a moving window was implemented, which makes it possible to carry out long time-scale and long scale length simulations by limiting the simulation box to a small region around the drive laser pulse, moving at the speed of light. The results of these simulations were qualitatively in agreement with our test particle simulations.

The second method that was explored relied on direct ponderomotive acceleration. In vacuum, energy exchange between a relativistic electron and a laser field is not possible if one considers only the linear force of the laser field. For ultra-intense laser pulses, however, the nonlinear or "ponderomotive" force can dominate the laser-electron interaction and a net energy exchange between the laser field and the electron is possible in vacuum. Preliminary analysis indicated that a properly shaped 50-femtosecond pulse of a Ti:Sapphire laser with  $\sim 1 \text{ J}$  energy focused at a gas target will accelerate  $\sim 10^6$  electrons to  $\sim 6 \text{ MeV}$  with  $\sim 0.01 \text{ mm-mrad}$  normalized emittance,  $\sim 0.5\%$  energy spread, and attosecond duration. Simulations were carried

out to determine the potential of this process and to design a well-optimized experiment. Also, to evaluate how space charge and finite energy spread affect the quality of these low-energy, high-density beams while propagating in vacuum, a particle tracking code was developed.

In the third technique we proposed obtaining  $\sim 400$  attosecond electron microbunches containing  $\sim 10^5$  electrons from a several-picoseconds long electron bunch supplied by a conventional linear accelerator. We considered obtaining either a sequence of  $\sim 103$  identical microbunches separated by  $\sim 1$  micron distance or a solitary microbunch.

Our studies have shown promising results confirming initial expectations. The next step is to do a real experiment, which we are planning now together with a group of physicists from the Advanced Test Facility at Brookhaven National Laboratory. A method for obtaining attosecond electron microbunches using a conventional linac was analyzed. The simulations indicate that the generation of pulses with a duration of 380 attoseconds [(Full Width at Half Maximum (FWHM))] should be possible.

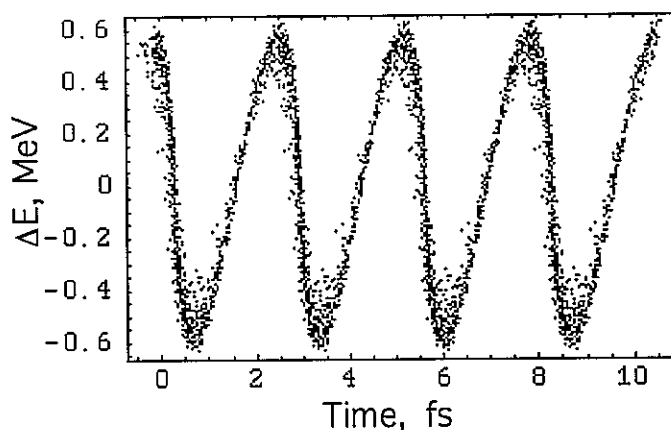


Figure 2: Energy modulation on a relativistic electron beam by co-propagating a laser beam with the electron beam through an undulator.

## Publications

A. Zholents, M. Zolotarev, and W. Wan, "Generation of Attosecond Electron Bunches," *2001 Particle Accelerator Conference*, Chicago, Illinois, (June 18-22, 2001)  
[http://pacwebserver.fnal.gov/papers/Friday/AM\\_Oral/FOA/C004.pdf](http://pacwebserver.fnal.gov/papers/Friday/AM_Oral/FOA/C004.pdf)

G. Fubiani, W. Leemans, and E. Esarey, "Studies of Space-Charge Effects in Ultrashort Electron Bunches," *Advanced Accelerator Concepts, 2000*, Santa Fe, New Mexico, **569**, 423-435 (2001).

R.E. Giacone, J.R. Cary, E. Esarey, W.P. Leemans, D. Bruhwiler, P. Mardahl, and J.P. Verboncoeur, "Simulations of Electron Injection into Plasma Wake Fields by Colliding Laser Pulses Using XOOPIC," *Proceedings from 2001 Particle Accelerator Conference*, 4022, Chicago, Illinois (June 18-22, 2001)  
[http://pacwebserver.fnal.gov/papers/Friday/AM\\_Poster/FP/AH149.pdf](http://pacwebserver.fnal.gov/papers/Friday/AM_Poster/FP/AH149.pdf)

---

## Compact Deuterium-Deuterium (D-D) Neutron Generator for Moderator Design and Biological Research

---

Principal Investigators: Ka-Ngo Leung, Gordon Wozniak, Rick Firestone, Eleanor Blakely, Steve Kahl, and Jasmina Vujic

Project No.: 00002

## Project Description

Several areas of basic research and applied neutron research require an intense source that is compact, inexpensive, and safe to operate. The current options for

neutron sources are reactors and high-energy accelerators. The plasma and ion source technology group of the Accelerator and Fusion Research Division has developed a unique technology that provides a sealed neutron tube producing a neutron flux that is about four orders of magnitude higher than current commercially available sources. The advantages of the D-D neutron generator are multiple: it only requires a low-energy accelerator; and it is portable, compact, and cheaper than other neutron sources. The availability of a high-intensity neutron source would open up new and exciting research possibilities, including medical and biological applications, material research, airport security, mine detection, non-proliferation technologies, and other radiographic applications.

The goal of this Laboratory Directed Research and Development (LDRD) project is to advance the developed concepts for a compact neutron generator to the final design, fabrication, and testing stage. We expect to be able to build a modest-sized neutron generator capable of producing D-D neutrons at  $10^{12}$  neutrons/second, and achieve several milestones in basic and applied testing.

## Accomplishments

During FY 2001, we have completed the design and fabrication of three compact neutron generators: a 2.5-centimeter-diameter axial generator, and a 5- and a 20-centimeter-diameter coaxial generator. All these generators have been tested for D-D neutron production. We first produced neutrons in March 2001 with a neutron output of  $3 \times 10^6$  neutrons per second by using a beam-driven titanium target arrangement. Recently, the neutron output flux has been increased to  $3 \times 10^7$  neutrons per second. The neutron output scales directly as the beam-on-target power as we anticipated. At present, the output is limited by the high voltage power supply. We are going to acquire a 120-kilovolt, 1-ampere, direct current power supply in FY 2002. This will enable us to generate D-D neutrons with flux as high as  $10^{12}$  neutrons per second.

Using the Monte Carlo N-Particle (MCNP) code, a simulation study was performed in the context of using 2.4 MeV D-D neutrons for the medical application of boron neutron capture therapy (BNCT), for Prompt Gamma Activation Analysis (PGAA) and for neutron scattering measurements for condensed matter applications. For the treatment of deep-seated brain tumors, it has been shown that 10-keV neutrons are optimal in order to maximize the treatment characteristics. Starting with D-D high energy neutrons, neutron energy spectra peaking around 10 keV could be obtained after moderation. The treatment time is about 80 minutes. Another simulation study has been carried out for applications requiring thermal or cold neutrons (such as PGAA and neutron scattering). It is found that both the moderated neutron spectrum and flux are very similar to that of the Los Alamos spallation neutron source

LANSCe (Los Alamos Neutron Science Center). The thermal neutron pulse width is about 17 microseconds.

## Prompt Gamma Activation Analysis

Initial experiments were completed that demonstrated prompt gamma activation analysis (PGAA) and neutron activation analysis (NAA) with the portable 2.5-centimeter-diameter neutron generator. These experiments were performed with the neutron generator operating at low duty cycle and producing about  $3 \times 10^6$  neutrons per second. The gamma spectra were recorded with a portable germanium detector. Different materials were irradiated inside a polyethylene moderator approximately 5 centimeters from the neutron generator and prompt and delayed gamma ray spectra were measured. The results confirm the validity of using a compact D-D neutron generator for PGAA/NAA analysis.

## Biology

In preparation of planned biological experiments using thermal neutrons, neutronics studies have been performed. Moderator and reflector configurations that maximize the thermal neutron flux at the sample position have been evaluated, and values for the required neutron yields from the generator tube have been established. The neutron fluxes that have been available thus far have been inadequate for the radiobiological studies planned, which involve the screening of the cell-killing effectiveness of new boron pharmaceuticals. These proposed studies have now been scheduled for completion during FY 2002. The present availability of neutrons, even at relatively low flux, has raised the possibility of responding to calls for proposals from Department of Energy (DOE) and National Aeronautics and Space Administration (NASA). Both agencies have an interest in the occupational risks associated with exposures to low doses of neutrons from work at power-generating reactors, and from work in space flight.

## Multi-Purpose Neutron Laboratory Concept

A multi-purpose neutron laboratory concept (see figure) based on a compact D-D neutron generator has been developed. The envisioned laboratory will benefit from the special advantages provided by a D-D neutron generator: small tube size, low neutron energy (2.45 MeV), modest shielding requirements ( $\sim 0.5$  meters), possible short distance between source and sample, variable pulse frequency (continuous to several kHz), and high flexibility by tailoring moderator and shielding structure to individual experiments. Neutronics simulations using the MCNP code were performed to optimize moderator configurations and to determine neutron beam properties. These studies were aimed at a wide variety of applications, ranging from optimizing the neutron field for prompt gamma activation analysis to designing neutron beams with small angular

divergence, short pulse width, and maximized neutron flux for scattering applications.

A D-D neutron generator with a neutron yield of  $10^{12}$  neutrons per second can serve as a research tool for small angle neutron scattering and reflectometry experiments and

as a training and teaching tool in the areas of neutron diffraction and inelastic scattering. It is also an excellent neutron source for activation analysis, radiography, and detector and moderator development.

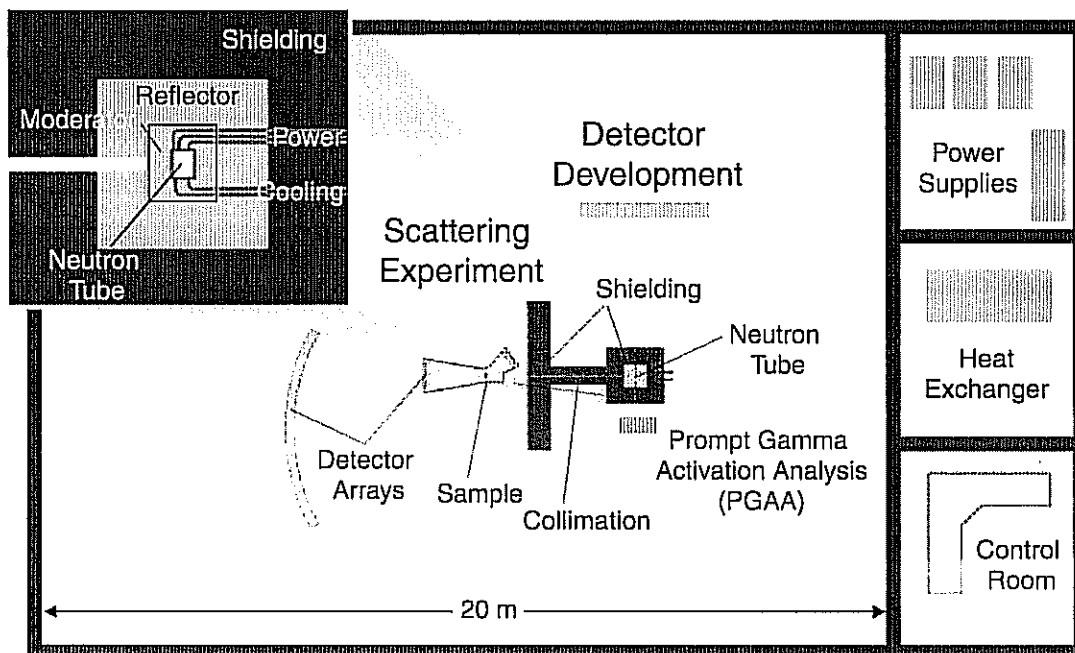


Figure 3: Conceptual design of multipurpose neutron laboratory.

## Publications

W.A. Barletta, "Compact Neutron Sources for Pure & Applied Science," *Proceedings of the Seminar on Planetary Emergency*, Sicily, Italy (August 19-24, 2001).

J. Reijonen, T.P. Lou, B. Tolmachoff, K.N. Leung, "High Flux Compact Neutron Generators," *Proceedings of the 7<sup>th</sup> International Conference on Applications of Nuclear Techniques* (July 21, 2001).

K.N. Leung, R.B. Firestone, T.P. Lou, J. Reijonen, and J. Vujic, "Compact Neutron Generators for Environmental Recovery Applications," *International Conference on Environmental Recovery of Yugoslavia* (September 27-30, 2001).

## Large-Scale Simulation of High-Intensity Beams in Circular Accelerators

Principal Investigators: Robert Ryne

Project No.: 01039

## Project Description

The goal of this project is to develop a capability to perform very high resolution simulations of intense beams in radiofrequency (rf) circular accelerators using parallel supercomputers. Such a capability does not currently exist. However, it is critical to the design of many next-generation accelerator programs of the Department of Energy (DOE), including future spallation neutron sources and proton drivers for neutrino factories and muon colliders. For such projects, minimizing beam loss is a key

issue, because excess losses will lead to high levels of radioactivity that can damage components, and that can hinder or prevent hands-on maintenance. The ability to predict phenomena such as beam halo formation and beam loss in high-intensity circular accelerators will have important consequences for the design and optimization of future machines, helping to reduce cost and risk while ensuring that performance requirements are met.

Our approach will use symplectic split-operator methods to combine the techniques of magnetic optics (including circular machine dynamics) with the techniques of parallel particle-in-cell simulation. Since magnetic optics codes usually use the arc length as the independent variable, a key issue for us will be the inclusion of certain transformations that are needed to compute the beam's space charge when the beam is located inside a bending magnet.

## Accomplishments

Features of the IMPACT (Integrated-Map and Particle Accelerator Tracking code) linac modeling code have been encapsulated and incorporated into the MaryLie Lie algebraic beam dynamics code. The code has been further enhanced so as to produce a single, coherent, parallel code for treating linear and circular accelerators both with and without space charge. This has been accomplished within a problem-solving environment that includes powerful fitting and optimization capabilities. Modifications and new capabilities include:

- Incorporation of the IMPACT model of rf accelerating cavities.
- Incorporation of IMPACT's Poisson solvers.
- Ability to model accelerating beams.
- Ability to use arbitrary "scaling" variables, since different variables are normally used to treat beams with/without acceleration.
- Ability to perform "automatic slicing" of thick beamline elements.
- Ability to automatically perform specified operations before/in the middle/after slices of elements. Such a capability can be used, for example, to plot trajectories and produce lattice function plots. Furthermore, the inclusion of space-charge effects based on a split-operator method is accomplished by making use of this new feature.

During the course of these modifications, the hybrid code was checked extensively against known analytical and numerical solutions, both with and without space charge.

However, a key motivation for this work was to enable a capability to accurately model very large amplitude, low-density beam halos for which existing benchmarks were not suitable. For this reason, new analytical and numerical work was performed in order to obtain a quasi-analytical test case, which had a very large amplitude tail, and which could be used to benchmark the new code. The test case involved derivation of the equations for a spherically symmetric, self-consistent, bi-thermal, stationary solution of the Vlasov/Poisson equations. The new distribution was generated numerically in such a way that 1% of the charge was located in the low-density tail that extended 8 standard deviations from the beam core. The MaryLie/IMPACT hybrid code was then used to propagate the beam and verify the stationary nature of the numerical distribution.

## Publications

R. Ryne, "Release Notes for MaryLie with Space Charge," (draft).

---

## ***A Source of Far-Infrared Radiation Based on a Material with an Optically Switched Permittivity***

---

Principal Investigators: Max Zolotarev, Robert Schoenlein, and Alexander Zholents

Project No.: 01005

## Project Description

The purpose of this work is the design and demonstration of a laser-assisted source for electromagnetic radiation in the far-infrared region based on a material with a high threshold for breakdown under electric field and optically switched permittivity. The goal is to reach ~1 gigavolt per meter (GV/m) of the radiation field near the surface of the material. This radiation field can be used for high-energy gain acceleration in a miniature, pencil-size accelerating structure—the electromagnetic wave undulator—and as an electron source with an ultra-low electron beam emittance. The long-term goal foresees the accelerator as a high-energy physics and/or a compact tabletop source of femtosecond x-ray pulses, utilizing self-amplified, spontaneous emission of electrons in interaction with the above-mentioned electromagnetic wave undulator.

This research will include three components: (1) development and characterization of a material capable of radiating a field of  $\sim 1$  GV/m, (2) study of the radiation mechanism based on the abrupt change in material permittivity, and (3) a proof-of-principle experiment. Theoretical and computational tools will be developed with an aim to design, model, and analyze a new source of far-infrared radiation based on an optically switched permittivity of the material. The proof-of-principle experiment will be conducted using a colored glass filter as a model of the desired material. Ways to design a new material will be explored.

## Accomplishments

The first year of this project had several highlights:

- Using the available 8kV pulser with a pulse length of 10 nanoseconds and 1kHz repetition rate, we were able to apply  $2 \times 10^6$  V/cm to the Schott Glass long pass filter GG475 without any evidence of an electrical discharge. This filter is made from a glass doped with cadmium sulfur (CdS) crystallites with the size distribution between 10 to 50 nanometers and with a
- semiconductor band gap of 2.8 eV. It makes it very suitable for a generation of THz pulses.
- To achieve this result, we tried out different insulator materials to suppress surface discharge between two electrodes. It turned out that the best result was obtained with an epoxy compound developed in cooperation with Applied Poleramics in Benicia with the help of Dr. Vorobyev.
- We designed and assembled a system for measuring of 100-femtosecond electric field pulses. In order to test this system, we developed a THz pulse generator based on gallium arsenic (GaAs).
- In order to be able to test different samples in fields up to  $2 \times 10^7$  V/cm we purchased from Russia a unique pulse generator capable of delivering 70-kV nanosecond pulses at 1 kHz repetition rate.
- This new generator has now been successfully tested and commissioned.

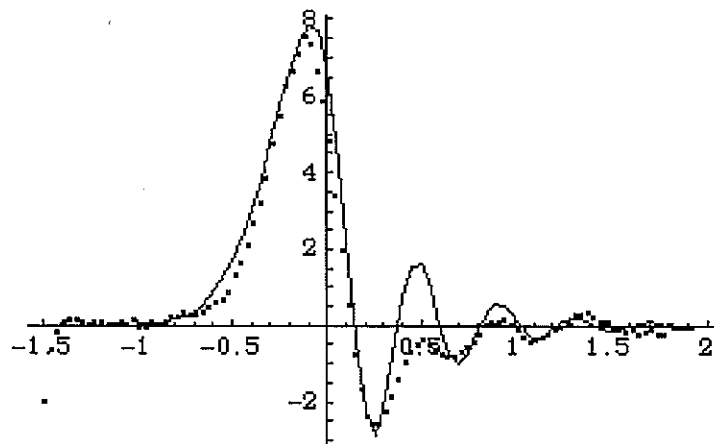


Figure 4: THz pulse measurement using electro-optical effect in zinc tellurium (ZnTe): dots are experiment and solid line is theory. Horizontal axis is time picoseconds, vertical axis is electric field in arbitrary units.





# Advanced Light Source Division

---

## ***Filling in a "Big Gap": A Novel Momentum-Resolved Inelastic Emission Soft X-Ray Spectrometer for the Study of Strongly Correlated Quantum Systems***

---

Principal Investigators: M. Zahid Hasan, Jonathan Denlinger, James Underwood, Zhi-xun Shen, and Zahid Hussain

Project No.: 01006

### **Project Description**

Despite extensive research efforts, the electronic structure of strongly correlated quantum systems continues to be a major class of unsolved problems in physics. The essential physics of these systems are believed to be dominated by the strong Coulomb interactions among relatively localized valence electron (d-electrons in the case of, for example, transition metal oxides). Well-developed momentum-resolved spectroscopies such as photoemission and neutron scattering cannot directly probe valence charge-charge correlation (fluctuation) spectrum in a momentum-resolved manner, as angle-resolved photoemission probes the single-particle occupied states and neutrons do not couple to the electron's charge directly. Optical Raman and infrared spectroscopies measure charge-fluctuation spectrum (occupied to unoccupied) but they are confined to the zone center ( $q \sim 0$ ) hence not momentum-tuned. A good understanding of momentum-resolved charge-charge correlation function is of paramount importance to gain insights into the charge-transport mechanisms in correlated systems. In addition, there is no momentum-resolved bulk spectroscopy to study the unoccupied states. To fill in this gap in electronic spectroscopies we are building a novel soft x-ray spectrometer that is 100 to 1000 times more efficient than previous designs.

Previous hard x-ray ( $\sim 10$  keV) resonant inelastic experiments at the transition metal K edges have demonstrated its applications for the study of momentum structure of the upper Hubbard band in cuprates. Exciting results have also been obtained regarding excitons in

correlated systems and their dispersion. However, the resonant scattering at the transition metal L (2,3) edges promise to be very spectacular and should also, according to the theory, have at least several orders of magnitude more scattering intensity in the inelastic channel. It would be invaluable to be able to look at the charge, lattice, or spin excitation in highly correlated materials with resolution limited only by the finite temperature of the sample.

### **Accomplishments**

Considerable progress has been made in designing a novel spectrograph that is optimized around Mn L-edge (500 to 700 eV) where it will focus on studying the charge excitations near the edge of the Mott gap (effective charge gap) in insulating manganites. The implementation of the following design characteristics has made this design very unique in providing a significant efficiency gain over the existing spectrographs:

- The instrument is slitless (thanks to high brightness of the Advanced Light Source). This requires a pre-focusing system to illuminate the sample with a spot size of about 5 microns in the vertical direction.
- The acceptance solid angle has been increased for collection of higher photon flux by incorporating a spherical pre-mirror.
- A blazed grating is optimized for the desired 640 eV and with nickel coating is used for higher efficiency.
- A back illuminated, two-dimensional, charge-coupled device (CCD) detector with high spatial resolution and oriented such that it maximizes the quantum efficiency of the detector.

We have already received the high-precision spherical pre mirror and blank substrate for ruling of the grating grooves. The order for spacing grating has been placed and we expect to receive the grating early 2002. The optical design of the spectrograph is completed. Mechanical design is in progress. A schematic of the spectrograph is shown below. An outstanding post doc has been hired and will start working on this project from January 2, 2002. We expect to start commissioning the unit from March 2002.

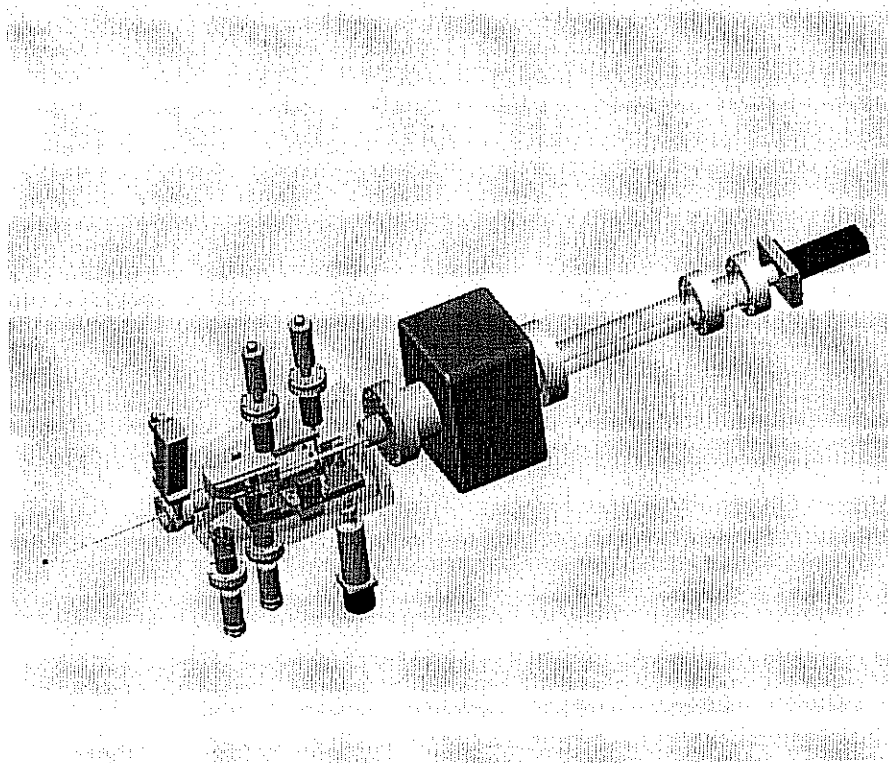


Figure 5: A schematic of the emission spectrograph.

## ***Investigation of Coherent Far-Infrared Synchrotron Radiation at the Advanced Light Source***

Principal Investigators: Michael Martin, John Byrd, Wayne McKinney, David Robin, Alexander Zholents, and Robert Schoenlein

Project No.: 01007

### **Project Description**

We proposed to search for super-radiant far-infrared synchrotron radiation at the Advanced Light Source (ALS). The overall goal is to understand enough of the accelerator physics underlying coherent synchrotron emission for a new far-infrared capability for the ALS. As a first crucial step, the details of how to produce stable, well-defined super-radiant emission from a storage ring needs to be understood.

Super-radiant, temporally coherent synchrotron radiation could significantly enhance the available photon flux at far-infrared wavelengths. At wavelengths longer than the electron bunch length in a storage ring, the individual electrons radiate in phase. This means that the radiative power in this long-wavelength regime is proportional to the square of the number of electrons, as opposed to the linear relationship that is well known with conventional (incoherent) synchrotron light sources. Even at low beam currents this flux enhancement is huge.

We will perform experiments to elucidate some of the underlying details of super-radiant synchrotron emission. The first will investigate coherent emission from the ALS electron bunches when the ring parameters are tuned for small bunch lengths or for instabilities that create micro bunching. The second set of experiments will look for coherent far-infrared light produced by a short electron bunch going through a bend-magnet in a linear accelerator system. A detailed comparison between existing theoretical predictions and the measured output from the two proposed experiments will then be done.

## Accomplishments

### Time Domain Measurements

As a first investigation into coherent far-infrared synchrotron radiation at the ALS, we placed a liquid helium-cooled Silicon Bolometer with integrated pre-amplifiers just outside a 20-millimeter diameter diamond window mounted in the "switchyard" at Beamline 1.4. A single extra mirror was inserted in the switchyard to direct the collimated beam through this window into the bolometer without disturbing the alignment of the infrared beamlines. The output of the detector was recorded by a digitizing oscilloscope. We observed large intensity bursts when the single bunch current was very high. The upper panel of the Figure shows a time trace of the bolometer output voltage for high-current single bunch operation. Although the bursts seemed to be quasi-random, at certain currents the bursts occurred within a periodic envelope. The rise time of the bursts was detector limited, while the fall time was approximately exponential with a time constant of  $\sim 310$  microseconds, probably due to the re-thermalization time of the detector. The threshold for the onset of bursts is approximately 7 mA at 1.5 GeV. The transition to bunching of the bursts within a super period occurs at about 27 mA at 1.5 GeV.

We verified that the coherent emission scales correctly with anticipated bunch densities causing instabilities and bursting by changing the electron beam energy, and the momentum compaction of the ALS ring.

### Spectral Measurements

By using a combination of filters with the above setup we determined the spectral content of the bursts was primarily below  $100\text{ cm}^{-1}$  (wavelengths longer than 100 microns). To make this more quantitative, we used the Fourier Transform Infrared (FTIR) spectrometer on Beamline 1.4.2 to measure the bursts that occur at the very top of the fill during regular two-bunch mode operations. By quickly averaging 100 interferograms over 50 seconds we integrated long enough to demonstrate the average spectral content of the bursts. Shown in the lower panel of the Figure is the measured intensity of the bursts as a function of wavelength, ratioed to the signal at low beam current. The bursts are peaked at  $\sim 27\text{ cm}^{-1}$ . This indicates a microbunching within the electron bunch having a period on the order of 400 microns (or approximately 30 times smaller than the normal ALS bunch length). The bursting was clearly dependent on the beam energy, with higher intensity at 1.5 GeV. This is to be expected, as the bunch length is proportional to  $E^{3/2}$ , implying higher peak currents at 1.5 GeV than 1.9 GeV, and therefore a greater tendency for instabilities and hence microbunching. Overall, the burst intensity dropped with decreasing beam

current, with the spectral content remaining essentially unchanged.

### Additional Measurements

We have begun additional measurements in collaboration with researchers at Jefferson Laboratory and Brookhaven National Laboratory to measure the coherent far-infrared emitted from a bend magnet in the Jefferson Lab energy-recovery linear accelerator based infrared free electron laser. Electron bunches at Jefferson Lab are typically a few hundred microns in length and should be emitting coherent far-infrared. Initial results show five orders-of-magnitude higher signal in the coherent emission compared to a conventional thermal far-infrared source.

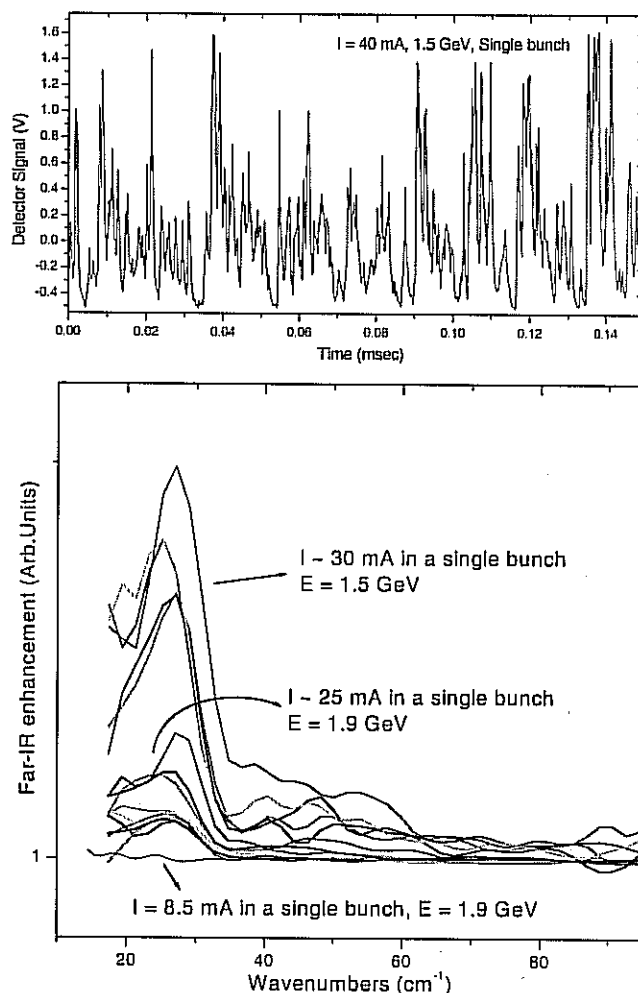


Figure 6: Upper Panel: Far-infrared detector signal versus time showing the bursting nature of the measured coherent far-infrared synchrotron emission. Lower Panel: Spectral content of the coherent far-infrared bursts. The signal is consistent coherent emission from a  $\sim 400$  micron long electron bunch length.

## Publications

M.C. Martin, J. Byrd, and W. McKinney, "Interesting Far-IR Bursts Measured at BL 1.4.2," *ALS Beamline Note* (February 26, 2001).

J.M. Byrd, A. Loftsdottir, M.C. Martin, and W.R. McKinney, "Observations of Bursting Coherent Far-Infrared Synchrotron Radiation in the ALS," in preparation for submission to *Nuclear Instrumentation and Methods A*.

G.L. Carr, M.C. Martin, W.R. McKinney, K. Jordan, G.R. Neil, and G.P. Williams, "Giant Relativistic Enhancement of Terahertz (THz) Radiation Yields 20 Watts of Average Power," in preparation for submission to *Science*.

---

## High-Pressure Research at the Advanced Light Source

---

Principal Investigators: Howard Padmore, Paul Alivisatos, and Raymond Jeanloz

Project No.: 01008

## Project Description

The goal of this work is to perform technical demonstrations using the Advanced Light Source (ALS) for high-pressure physics, geophysics and chemistry. The research will be centered on understanding structural and chemical dynamics in nanocrystalline materials, and examining the chemical reactions that occur at high pressure and temperature inside the Earth and planets. This work will also strengthen the support of the high-pressure research community for a dedicated superbend beamline for this type of research at the ALS.

Our approach is to use an existing beamline at the ALS and equip it with the necessary hardware to perform high pressure and temperature experiments, using the probes of x-ray absorption spectroscopy to deduce chemical state, large-angle scattering to determine atomic packing, and small-angle scattering to determine the shape of molecular assemblies. This is being made possible by the move of another research group onto a dedicated beamline, therefore freeing space and beamtime. We also intend to make use of the adjoining laser lab for a high-power laser for sample heating, and we already have much of the infrastructure, such as a ruby fluorescence system for pressure measurement and diamond anvil cells for application of

pressure. A particular thrust of this program will be to develop x-ray spectroscopy at energies < 13 keV, requiring thin diamonds or the use of beryllium gaskets in transverse mode.

## Accomplishments

We have established a capability for high-pressure diffraction experiments on Beamline 7.3.3. This has involved:

- The development, installation, and commissioning of a novel cryo-cooled monochromator.
- The development of an alignable bench with appropriate beam conditioning apparatus.
- Demonstration of high quality diffraction patterns from crystalline systems.
- Demonstration of both wide- and small-angle scattering experiments from nano-particles.
- Studies of important geological processes.
- Studies of a range of nano-particle systems.

Currently, it is possible to collect high quality diffraction data in a few minutes from nano-particle systems.

We have successfully demonstrated that the ALS is a suitable source for high-pressure diffraction studies and made major contributions to a number of University of California, Berkeley-based research programs. The scientific highlights include the discovery and characterization of a new phase of oxygen and a study of the effect of particle size and solvent conditions on the gamma to alpha transition of iron oxide nanocrystals.

This work has already led to some follow-on funding for a dedicated high-pressure facility on one of the new superbends at the ALS. Additional funds are also being sought for a second high-pressure station on a superbend beamline.

We are currently adding increased automation with the aim at higher levels of scientific throughput. This involves adding an *in-situ* pressure measurement system that will decrease the time required per pressure point from about thirty minutes to about five minutes.

We now plan to demonstrate the usefulness of the ALS for high-pressure spectroscopic measurements at low energy edges, for example iron and manganese, using diamond anvils laser drilled to lower absorption, and to carry out small-angle scattering studies of nano-particle transformations.

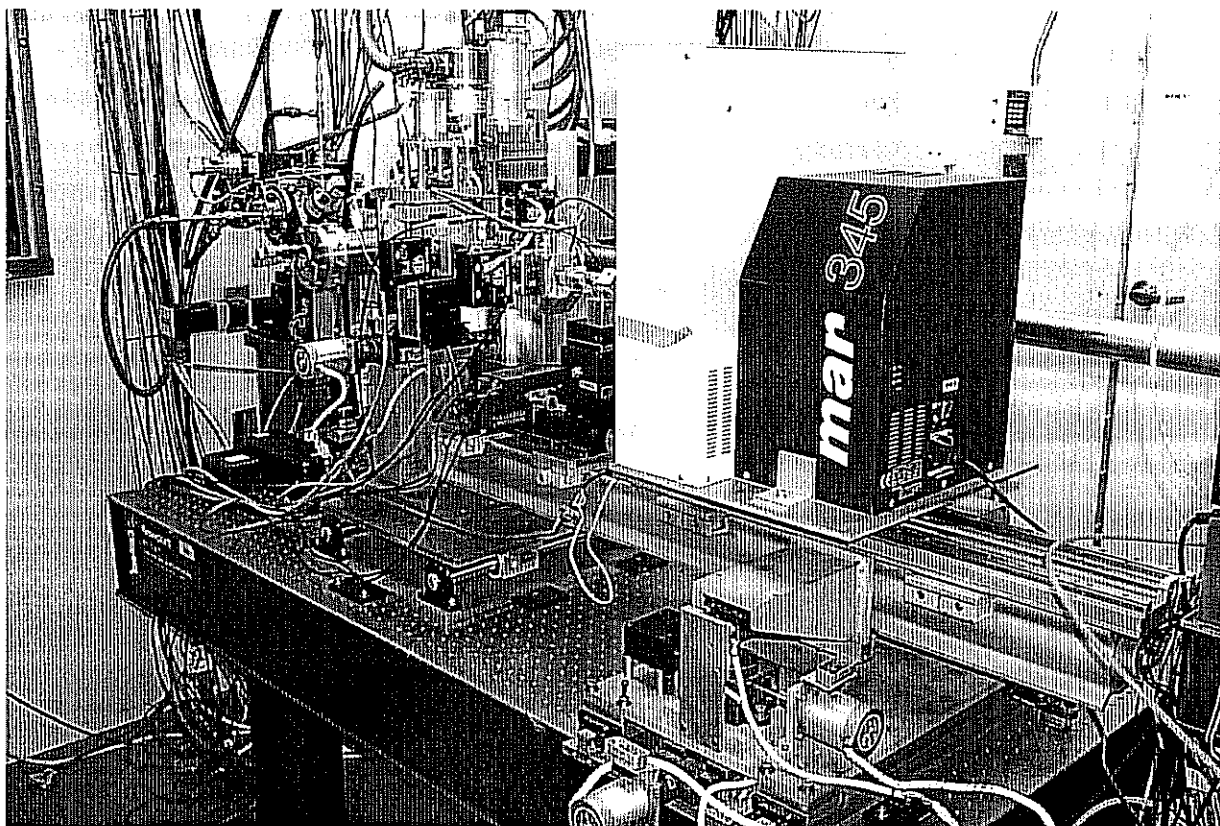


Figure 7: High-pressure experimental set-up on Beamline 7.3.3.

## Publications

S.G. Prilliman, J. Rockenburger, S.M. Clark, and A.P. Alivisatos, "Size Dependent Transitions in Gamma-Fe<sub>2</sub>O<sub>3</sub> Nano-Crystals," in preparation for *Physical Review Letters*.

L.R. Benedetti, W.R. Panero, S. Akber, K.K.M. Lee, S.M. Clark, and R. Jeanloz, "Oxygen in the Solar System: Dissociation of H<sub>2</sub>O to Hydrogen and a New Phase of Oxygen at High Pressure and Temperatures," in preparation for *Science*.

S.M. Clark, R.S. Celestre, A.A. MacDowell, E.A. Domning, A.C. Thompson, H.A. Padmore, S.G. Prilliman, A.P. Alivisatos, and R. Jeanloz, "A New High-Pressure Diffraction Facility at the Advanced Light Source," in preparation for *Synchrotron Radiation News*.

---

## Orbital Ordering in Colossal Magnetoresistance

---

Principal Investigators: John Spence

Project No.: 01009

## Project Description

The useful electronic properties (magnetoresistance, superconductivity) of strongly correlated materials are known to depend on a subtle temperature and doping-dependant competition between charge, spin, and orbital ordering. We have recently demonstrated a new method for real space imaging of orbital holes (charge densities) and electron transfer. We aim to use this to observe orbital ordering in LaMnO<sub>3</sub> in real space, to measure one-electron d-orbital hybridization coefficients, electron transfer, covalency, and to test, by comparison with FLAPW/LDA/GGA/Hubbard-U calculations, various many-electron schemes now used to predict ground state

properties of new materials. Extensions to x-ray diffuse scattering from striped phases and Jahn Teller (JT) polarons in  $\text{La}(1-x)\text{Ca}(x)\text{MnO}_3$  are envisaged.

Real-space mapping of charge ordering and bonds requires <1% accuracy in structure factor (Fg) measurement, but has been hampered by extinction, the need for large single crystals, and weak reflections. We have shown that the Mott-Bethe relation between x-ray and electron Fg solves this problem, and that a combination of x-ray and electron Fg measurements provides a unique tool for the quantification of charge and orbital ordering. Unavoidable twinning has prevented previous work on  $\text{LaMnO}_3$ ; we have now grown (at Bell Labs) single crystals with one-micron twins. Post-doc H. He arrived in September 2001 for this project. The existing Bruker D8 small Kappa goniometer and SMART APEX CCD and software are currently being installed on Beamline 11.3.1., together with Oxford Cryostream cooling, by H. He and A. Thomson. Extensions of this work (with J. Patel) to the new boride superconductors are straightforward.

## Accomplishments

Beamline 11.3 at the Advanced Light Source (ALS) has been built to enable us to solve complex crystal structures, and to enable us to extract highly precise bonding information from the details of the charge distribution around individual atoms. A special feature of this system will be the ability to work with very small crystals, down to a few microns in size. The beamline development has been pursued under this Laboratory Directed Research and Development (LDRD) project, and in the next few months we expect to begin taking crystallographic data associated with the charge density mapping techniques that we plan to apply to colossal magnetoresistance (CMR) materials. The beamline monochromatizes and focuses x-rays from the ALS, and directs them into an end station enclosure. The monochromator and x-ray optics are located inside the shield wall and are complete. Beam has been extracted into the end station area, and a focused beam has been observed on a phosphor screen. With completion of the x-ray hutch, the beamline will be fully optimized and commissioned. The charge density "orbital imaging" applied to CMR materials can then be started.

Due to the late commissioning of Beamline 11.3.1, the hiring of Post-Doc H. He from the Max Planck Institute, Stuttgart, Germany was delayed. He has since been working with A. Thomson at the ALS to install the goniometer, cooling device, and detectors for this experimental work. Crystals with large (e.g. one micron) twins have been grown. The methods developed for electron microscopy sample preparation are being used to make extremely small samples (a few microns or less in diameter) for the x-ray work. The electron microscopy

component of this project is underway at Arizona State University (Dr. B. Jiang), and several structure factors have been measured for  $\text{LaMnO}_3$ . Meanwhile, the possibility has arisen to collaborate with Dr. J. Patel on a charge-density study of the new boride superconductor  $\text{MgB}_2$ , using the same methods applied to  $\text{LaMnO}_3$ . Patel has very small single crystals, grown in Korea, suitable for both electron and x-ray work.

## Publications

J.C.H. Spence. "Charge Transfer in Mott Insulators," presentation at Gordon Conference on Charge, Spin and Momentum Densities (August 2001).

B. Jiang, J. Zuo, Q. Chen and J. Spence, "Orbital Ordering in  $\text{LaMnO}_3$  and its Effect on Structure Factors," *Acta Crystallographica*, **A58**, 4-11 (2002).

J.C.H. Spence, M. Howells, L.D. Marks, and J. Miao, "Lensless Imaging: A Workshop on 'New Approaches to the Phase Problem for Non-Periodic Objects,'" proceedings published in *Ultramicroscopy*, **90**, 1-6 (2001).

J.C.H. Spence, "Holography of Atoms," *Nature*, **410**, 1037 (2001).

M.R. Howells, C. Jacobsen, S. Marchesini, S. Miller, J. Spence, and U. Weierstall, "Toward a Practical X-Ray Fourier Holography at High Resolution," *Nuclear Instruments and Methods*, **A467**, 864-867 (2001).

J.C.H. Spence, "Workshop on Lensless Imaging," *Synchrotron Radiation News*, **14**, 6, 21-23 (Fall 2001).

## Chemical Sciences Division

---

### ***Laser-Assisted Photoabsorption of X-Rays by Atoms and Feasibility of a Femtosecond X-Ray Atomic Switch***

---

Principal Investigators: Ali Belkacem, Thornton Glover, and Michael Prior

Project No.: 00005

#### **Project Description**

An important effect resulting from dressing an atom with a high-intensity laser is the change of its atomic structure. A combination of high-intensity laser and high brightness of a synchrotron radiation source, such as the Advanced Light Source (ALS), offers a unique opportunity for probing inner-shell electrons and, in the process, opening the prospect of a new type of sub-picosecond x-ray atomic switch in the several keV energy-range. The study of the excitation of inner-shell electrons—the underlying process on which the “atomic” switch concept is based—requires a high degree of synchronization between the laser beam and a tunable x-ray beam that has a high energy resolution. This stringent requirement has limited the number of such studies reported in the literature, despite the critical role that inner-shell excitation processes play in many physical concepts including the x-ray laser. The state-of-the-art facilities at the ALS, in particular the new Beamline 5.3.1, are uniquely suited for such studies.

An x-ray, with several keV energy, impinging on a high-Z atom will interact essentially with core electrons. The absorption cross section of the x-ray by the atom can jump by an order-of-magnitude when the energy is tuned from below the K-absorption edge to above the K-absorption edge. Both initial and final state energy levels contain a sizable contribution from the strong electron-electron correlation of the multi-electron system. The application of a high-intensity laser field to a multi-electron atom adds another component. The structure of the laser-dressed atom will differ from that of the same atom in a field-free space as a result of two mechanisms:

- The electric field related to the short-pulse laser induces a linear and quadratic stark shift of the energy levels. This effect is larger for the outer atomic shells and continuum states.

- The outer electrons, strongly driven by the laser field, will rearrange and, in some cases, will move away from the ion core, decreasing the effect of their screening on the inner-shell. Consequently, the binding energy of the core-electrons will differ from that of the field-free atom. In the limit of ionization of the M-shell for argon, for example, the binding energy of the K-shell will sink in the Coulomb potential by 170 eV.

In the first stage of this project, we will use the laser facilities already existing at the ALS on Beamline 7.3.1.2 to develop the diagnostic tools and endstation that are needed for the experiment. In the second stage, we will move the apparatus to Beamline 5.3.1 to make use of the tunable femtosecond x-rays, a unique facility that is currently being built at the ALS.

#### **Accomplishments**

During this fiscal year we ran the first experiments on Beamline 5.3.1 both in the static mode (x-ray without laser) and time dependent mode (x-ray with laser). Since the femtosecond x-rays are not yet available on Beamline 5.3.1 we used the 50-picosecond long x-ray pulses.

One of the first measurements we did with these relatively long x-ray pulses was the study in the time domain of the relaxation of an x-ray induced K-vacancy of argon. We measured the ion charge state distribution of argon as the x-ray energy is varied across the K-edge. We found that even when the x-ray energy is tuned above the K-edge by several electron volts the charge state distribution of the argon ions does not change. This change in the ion charge distribution that one intuitively would expect to occur right above edge doesn't start occurring until over 10 eV above edge. This is an indication of a post-collision interaction in which the photoelectron is recaptured by the core ion that has in the process shed, very quickly, some of its inner electrons through an Auger process. This post-collision interaction process in the static mode has already been observed by Wayne Stolte and his group at the ALS. We set up the experiment to study this process in the time domain and made a measurement of the argon charge state distribution and the electron yield with laser-on and laser-off, where the laser is overlapped in time (100 picoseconds) and space (100 microns) with the camshaft pulse of the ALS x-ray pulse train. We found a very unexpected time dependence, in the nanosecond time scale, of the electron yield. This electron yield is correlated with the production of high charge state argon ions. This very unique set of data is currently being analyzed. In our current thinking, this

measurement would indicate that a sizable number of highly excited metastable argon atoms, with several electrons in excited states, survive on a nanosecond time scale. These highly excited metastable argon atoms are created during the process of relaxation of argon with a K-vacancy. The short laser pulse interacts very efficiently with this highly excited argon atom, accelerating its autoionization into highly charged ions.

These first successful experimental runs on Beamline 5.3.1 using a combination of femtosecond laser and x-rays show that we are able to overcome several problems associated with the mismatch of the kilohertz laser and half gigahertz ALS repetition rates. The main problem that we had to address is the synchronization between the femtosecond laser and the ALS x-rays. The ALS ring consists of electron buckets spaced by two nanoseconds. In the ring's normal running mode, a gap of several tens of nanoseconds is left empty with the exception of one filled electron bucket located somewhere in the middle of this gap. The position of this isolated bucket (camshaft) and the width of the empty gap are changed according to users requirements. We used microchannel plate detectors with very high extraction fields to resolve inner-shell ionization events generated by the x-rays of the camshaft bucket. We also designed a very elaborate electronic event-by-event read out that selects and displays camshaft events with and without overlap with the laser pulse. This detection technique kept the background from accidental events to a very low rate. This design also removes normalization problems associated with the change of the x-ray intensity with time. Another major challenge associated with gas phase experiments that we had to overcome is the design of a setup that allows a very high-density gas target while the detectors are kept in very high vacuum.

With follow-on funding from Office of Science/Basic Energy Sciences, we are currently changing the setup to include a three-dimensional imaging detection technique. This technique will allow us to study the modification of x-ray absorption induced by laser-dressing of argon using femtosecond x-rays.

---

## ***Development of a Neutral Molecule Synchrotron Storage Ring***

---

Principal Investigators: Harvey Gould and Hiroshi Nishimura

Project No.: 01010

### **Project Description**

Our goal is to determine the feasibility and practicality of constructing molecular synchrotrons to store, slow, and cool neutral polar molecules in a way that makes it easy to use them in novel new experiments, including the formation of molecular Bose-Einstein condensates.

Laser slowing, cooling, and trapping, have not been effective for molecules because the closely spaced (vibrational and rotational) levels prevent efficient cycling transitions. This makes it possible to apply accelerator and specifically synchrotron physics and technology to polar molecules. Molecular synchrotrons have the potential to be the first widely applicable and effective method to store, slow, and cool molecules in vacuum.

Static and dynamic traps have been considered for trapping molecules. The molecular synchrotron is effectively a dynamic (rf) two-dimensional trap. It has fewer constraints than a three-dimensional trap or a static trap. A molecular synchrotron's beam energy is independent of temperature and can be varied over several orders of magnitude. It preserves a beam geometry for experiments, can have field-free regions, and can be scaled to accept molecules with large kinetic energies. It is well suited for evaporative cooling of the molecules. It may be possible to use a ring as an accumulator to increase the stored intensity.

### **Accomplishments**

We have modeled a tabletop storage ring for slow polar molecules. Simulations show that the ring has large transverse acceptance and will hold the molecules for over 1000 turns. The model storage ring also has straight sections for injection and bunching or cooling the molecules and for experiments.

We have invented new molecular beam focusing lenses, needed for a molecular synchrotron, which can focus molecules in any rotational state. We have modeled a beamline, from source to final focus, using these new lenses and demonstrated that it can transport intense beams of



neutral polar molecules over extremely long distances and focus them.

## **Publications**

J.G. Kalnins, G. Lambertson, and H. Gould, "Improved Alternating Gradient Transport and Focusing of Neutral Molecules," submitted to *Review of Scientific Instruments*.

Manuscript in preparation to be submitted to *Physical Review Letters*, LBNL-49088 (Draft).

---

## **Photoionization Experiments on Atoms and Molecules Adsorbed onto Helium Droplets**

---

Principal Investigators: Daniel Neumark

Project No.: 01011

## **Project Description**

This project will perform photoionization and photoelectron spectroscopy experiments on atoms and molecules adsorbed onto large helium droplets ( $\sim 10^4$  atoms) in order to understand how these fundamental and well-understood gas phase processes are altered in the environment of a helium droplet. A helium droplet source will be constructed and used on Endstation 3 of the Chemical Dynamics Beamline at the Advanced Light Source. Photoionization mass spectrometry and photoelectron spectroscopy experiments will be performed using the high-throughput vacuum ultraviolet monochromator on this endstation.

## **Accomplishments**

During the past year, a helium-droplet molecular beam source was constructed and installed on Endstation 3 of the Chemical Dynamics Beamline, and first results were taken for pure helium nanodroplets consisting of about  $10^4$  atoms. We measured the total electron yield as a function of photon energy in the range of 20 to 24 eV. The spectra show sharp resonances corresponding to atomic helium excitation superimposed on a structured background. We also measured photoelectron spectra at a series of photon energies, both below and above the ionization potential of atomic helium. We observed photoelectron spectra clearly associated with the droplets; the electron kinetic energy

distribution peaks near zero kinetic energy and drops off monotonically toward higher electron kinetic energy.

Our plans for the coming year will focus on refining the above measurements, collaborating with theorists at University of California, Berkeley and Berkeley Lab to understand them, and to begin performing experiments on "doped" helium droplets containing a single argon atom or small molecule ( $H_2$ , NO).

---

## **Study of Radionuclide-Bacterial Interaction Mechanisms**

---

Principal Investigators: Heino Nitsche and Petra Panak

Project No.: 99006

## **Project Description**

This project is to determine the influence of microorganisms on radionuclide transport in the environment. A quantitative and mechanistic understanding will be developed of radionuclide interaction, in particular of the actinides uranium and plutonium, with aerobic soil bacteria. The information from this study is important for improving reactive transport models predicting the movement of actinides at contaminated sites and assessing the risk of potential nuclear waste disposal sites. Furthermore, the results will be transferable to possible biotechnology processes for stabilization and remediation of radionuclide-contaminated sites. The study is interdisciplinary and combines actinide chemistry, microbiology, and molecular environmental science.

The production and testing of nuclear weapons, nuclear reactor accidents, and accidents during the transport of nuclear weapons have caused significant environmental contamination with radionuclides. Due to their long half-lives and high chemical and radiotoxicity, the actinides pose a significant threat to human health. Their migration behavior is controlled by a variety of complex chemical and geochemical reactions such as solubility; sorption on the geo-medium; redox reactions; hydrolysis; and complexation reactions with inorganic, organic, and biological complexing agents. In addition, microorganisms can strongly influence the actinides' transport behavior by both direct interaction (biosorption, bioaccumulation, oxidation, and reduction reactions) and indirect interaction (change of pH and redox potential), thus immobilizing or mobilizing the radioactive contaminant. Very little information is

available about these processes. Our research focuses on the interaction of aerobic bacteria that are present in the upper soil layers with plutonium and uranium.

## Accomplishments

In the last year of this three-year project, we centered our research on the interaction of *Bacillus sphaericus* with hexavalent actinides as a function of age and status of the cells. Sorption studies with U(VI) and Pu(VI) provide information on the amount of U taken up by living cells, spores, and dead and decomposed cells. We used time-resolved laser fluorescence spectroscopy (TRLFS) and x-ray absorption spectroscopy [extended x-ray absorption fine structure (EXAFS) and x-ray absorption near-edge structure spectroscopy (XANES)] to characterize the complexes formed. Both methods are capable of differentiating between organo-phosphate complexes with phosphate groups on the cell surface and U(VI)/Pu(VI) - phosphate precipitates.

Sorption data of U(VI) and Pu(VI) for *Bacillus sphaericus* (vegetative cells, spores, heat-killed cells, and decomposed cells) as a function of biomass at pH 5 are presented in Figure 8. For a better comparison, the results were normalized to the dry weight of the bacteria. The relative biosorption efficiency decreased with increasing biomass concentrations. This is due to an increased agglomeration of the bacteria at higher biomass concentrations, which was observed by microscopy. These results have shown that it is very important to examine the sorption behavior of different strains at various biomass concentrations because different strains can display different sorption behavior with increasing or decreasing biomass concentrations. The spores of *Bacillus sphaericus* showed a much higher biosorption compared to the vegetative cells at low biomass concentration. Vegetative and heat-killed cells showed an almost identical sorption behavior. Decomposed cells removed a significantly higher amount of U(VI) compared to vegetative cells. More U(VI) than Pu(VI) was accumulated under the same conditions. Whereas U(VI) was stable, Pu(VI) was partly reduced to Pu(V).

To obtain information on the reversibility and the binding strength of the bacterial complexes, we extracted the cell-bound U and Pu with 0.01 moles per liter (M) ethylenediaminetetraacetic acid (EDTA) solution (pH 5). For all biomass concentrations, between 80 and 95 % of the U and Pu was released from the cells. We observed no significant differences of extraction behavior between different strains or between the vegetative cells and the spores. These results show that the process is reversible and confirms the formation of surface complexes with functional groups of the cell surface. The bacterial complexes are less stable than the U and Pu EDTA-complexes.

*Bacillus sphaericus* forms strong surface complexes with aqueous U(VI) and Pu(VI). Mainly phosphate groups on the cell surface are involved in Pu complexation. An average number of phosphate groups of 0.89 mmol/g dry weight for the strain used in our experiments, but not all phosphate groups determined by acid/base titration are available for binding of the Pu.

We used EXAFS spectroscopy to determine the structural parameters of the U(VI) and Pu(VI) bacterial complexes. Our results show that U(VI) and Pu(VI) are primarily bound to phosphate groups on the cell surface. No carboxylate complexation could be detected.

Studies on the interaction of U(VI) with living cells, spores, heat-killed cells, and decomposed cells of *Bacillus sphaericus* by time-resolved laser fluorescence spectroscopy show that different phosphate complexes are formed. Using fresh intact cells, the U(VI) is bound to organo-phosphate groups of the bacterial cells. With increasing age of the samples, the cells begin to lyse. During this process, inorganic phosphate is released and a uranyl phosphate precipitate is formed. The characterization of the released phosphate species by Raman spectroscopy and the uranyl phosphate complexes by time-resolved laser-induced fluorescence spectroscopy (TRLFS) has shown that the bacteria release  $\text{H}_2\text{PO}_4^-$  during decomposition which leads to the formation of  $\text{UO}_2(\text{H}_2\text{PO}_4)_2$ .

EXAFS spectroscopy of the Pu(VI) complex with *Bacillus sphaericus* confirmed these results. The analysis of the EXAFS data not only provides information on the bond lengths and the coordination number of the axial and equatorial oxygens and the phosphorus atoms, it also shows that the Pu(VI) in a six-hour sample is not precipitated as  $\text{PuO}_2(\text{H}_2\text{PO}_4)_2$  by potentially released phosphate. The feature of the EXAFS spectra and the corresponding Fourier transforms of the bacterial complex differ significantly from those of the  $\text{PuO}_2(\text{H}_2\text{PO}_4)_2$  precipitate. Furthermore, the bond lengths between the Pu(VI) and the equatorial oxygen as well as the phosphorus atoms are longer in the bacterial phosphate complex than in the Pu(VI) dihydrogen phosphate precipitate. The number of phosphorus atoms also decreases. In agreement with the results of U(VI), we conclude that Pu(VI) is bound to organo phosphate groups of the cell surface. Differentiation between complexes formed by interaction with bacteria is very important for studies in natural environmental systems where not only living metabolizing cells but also dead and even decomposed cells are present.

Contrary to U(VI), which is stable under our experimental conditions, Pu(VI) is reduced with increasing contact time to Pu(V) in the presence of aerobic soil bacteria. We investigated the oxidation state distribution of the Pu associated with the bacteria by fitting the XANES spectra

using Pu(IV), Pu(V) and Pu(VI) reference spectra. The results agree well with the oxidation state distribution obtained by solvent extraction. After one month of contact time, about 16% of Pu(IV) was found to be bound to the bacteria. This is not an effect of further microbial reduction, but merely due to the disproportionation of Pu(V) and the possible autoreduction of Pu(VI). Pu controls without the bacteria present showed also reduction from Pu(V) to Pu(IV).

Understanding the different kinds of interaction with *Bacillus sphaericus* as a function of the status and the age

of the samples is very important to better understand natural systems, where living cells, spores, dead and also decomposed cells are present. Detailed information on the main interaction with different kinds of cells of *Bacillus sphaericus* and their decomposition products will enable a better prediction on a possible impact of bacteria on the transport of actinides and can be a basis for developing new remediation strategies using bacteria for immobilization of actinides in waste deposits.

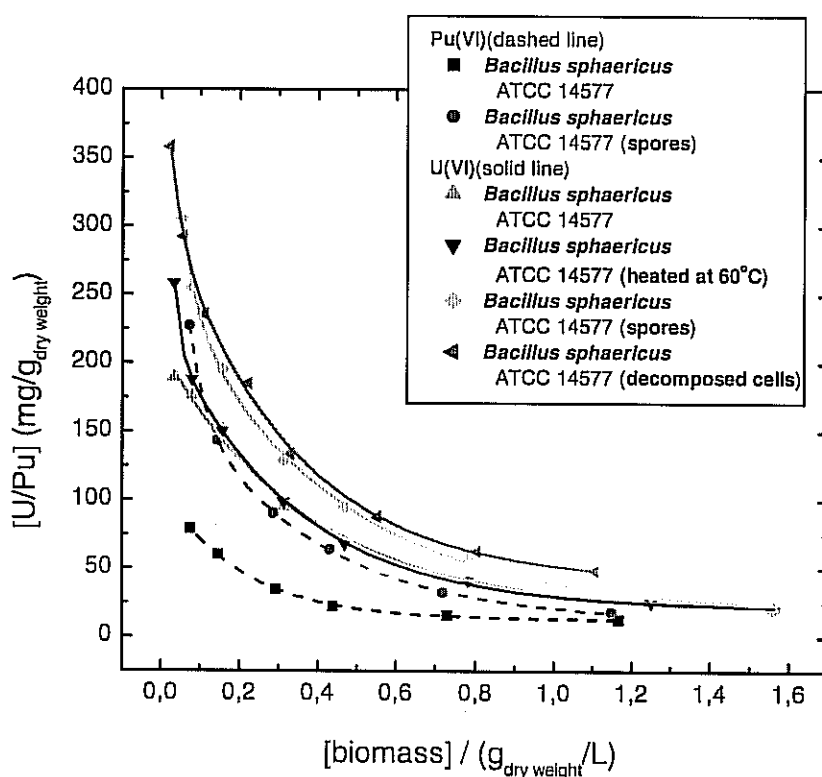


Figure 8: Uranium(VI) biosorption of vegetative cells, heat killed cells, spores, and decomposed cells of *Bacillus sphaericus* ATCC 14577 at pH 4.5 as a function of the biomass concentration in comparison to the Pu(VI) biosorption of vegetative cells and spores. The results are normalized to the dry weight of the bacteria.

## Publications

P.J. Panak, and H. Nitsche, Interaction of Aerobic Soil Bacteria with Plutonium(VI); *Radiochimica Acta*, **89**, **8**, 499-504 (2001).

P.J. Panak, R. Knopp, C.H. Booth, and H. Nitsche, "Spectroscopic Studies on the Interaction of U(VI) with *Bacillus sphaericus*," submitted to *Radiochimica Acta*.

P.J. Panak, C.H. Booth, D.L. Caulder, J.J. Bucher, D.K. Shuh, and H. Nitsche, "X-Ray Absorption Fine Structure Spectroscopy of Plutonium Complexes with *Bacillus sphaericus*," *Radiochimica Acta* (accepted).

P.J. Panak, R. Knopp, and C.H. Booth, "Interaction of Aerobic Soil Bacteria with Hexavalent Actinides (Pu(VI) and U(VI))," poster presentation at the 8<sup>th</sup> International Conference on "Chemistry and Migration Behavior of Actinides and Fission Products in the Geosphere (Migration 01)," Bregenz, Austria, (September 16-21, 2001).

R. Knopp, P.J. Panak, L.A. Wray, N. Renninger, J.D. Keasling, and H. Nitsche, "Investigation of Interactions of U(VI) with Bacteria by Laser Spectroscopic Methods," presented at the 8<sup>th</sup> International Conference on "Chemistry and Migration Behavior of Actinides and Fission Products in the Geosphere (Migration 01)," Bregenz, Austria, (September 16-21, 2001) (LBNL-47658 Abs.).

P.J. Panak, R. Knopp, C.H. Booth, and H. Nitsche, "Interaction of Aerobic Soil Bacteria with Hexavalent Actinides (Pu(VI) and U(VI))," presented at the 221<sup>st</sup> ACS National Meeting, San Diego, California, (April 1-5, 2001) (LBNL-47136 Abs.).

R. Knopp, P. Panak, and H. Nitsche, "Laser-Based Methods for the Investigation of Actinide Interactions with Colloids and Bacteria in Aqueous Environments," presented at the 221<sup>st</sup> ACS National Meeting, San Diego, California, (April 1-5, 2001) (LBNL-47138 Abs.).

P. Panak and H. Nitsche, "Plutonium and Aerobic Soil Bacteria," *LBNL-47060 Abs.*, presented at the 2000 International Chemical Congress of Pacific Basin Societies, Honolulu, Hawaii, (December 14-19, 2000).

R. Knopp, P. Panak, and H. Nitsche Laser Spectroscopic Interfacial Studies of Actinides on Bacteria and Mineral Surfaces, , Poster Presentation, Heavy Element Contractors' Meeting, Argonne National Laboratory, Argonne, IL, November 20-21, 2000. (LBNL-47076 Abs.)

# Computing Sciences

---

## High-Precision Arithmetic and Applications

---

Principal Investigators: David H. Bailey and Xiaoye Li

Project No.: 99007

### Project Description

For a growing number of investigators in mathematics, physics, and engineering, the standard computer hardware arithmetic typically found on today's computer systems (IEEE 64-bit format, accurate to approximately 16 digits) is no longer adequate. Some problems require only twice this amount ("double-double" precision), while others require substantially more, sometimes the equivalent of hundreds or thousands of decimal digits. High-precision arithmetic of this sort is typically performed by utilizing software packages, such as one that has been developed by the Principal Investigator.

One of the more interesting applications of very high-precision arithmetic is the discovery of new mathematical identities. Perhaps the most remarkable example of this is a new formula for the mathematical constant  $\pi = 3.14159\dots$ , which was discovered recently. This new formula has the surprising property that it permits one to directly determine the  $n$ -th binary or base-16 digit of  $\pi$ —without calculating any of the first  $n-1$  digits—by means of a simple algorithm that can easily run on a personal computer. Numerous other formulas of this sort have subsequently been discovered. It now appears that these results have more than recreational value—they have important implications for some centuries-old mathematical questions.

Other applications of high-precision arithmetic to be explored in this activity include finding relationships between constants that arise in quantum field theory, the study of vortex roll-up phenomena in fluid dynamics, and the stabilization of climate modeling calculations.

This activity proposed to develop some new, faster high-precision arithmetic software, together with high-level language bindings, so that these routines can be accessed from a user's Fortran and/or C programs with only minor modifications. Some continuing studies in applications of this software were also proposed, including research in physics, applied mathematics, and pure mathematics.

### Accomplishments

With the assistance of Brandon Thompson, an undergraduate student at University of California (UC), Berkeley, a new arbitrary precision computation package was completed. This software library includes routines to perform all of the standard arithmetic functions to an arbitrarily high level of numeric precision. There is one set of routines for "normal" high precision, namely 50 to 1000 decimal-digit accuracy. For requirements above this level, a separate set of routines, which employ advanced techniques such as fast Fourier transform (FFT)-based multiplication, have been developed.

In conjunction with the above, Sherry Li has completed a high-level language facility in C++, which permits the arbitrary precision library to be used from ordinary C++ programs, with only minimal programming effort. A related effort to develop a high-level Fortran-90 facility is not yet complete, and will be completed as part of a new Laboratory Directed Research and Development (LDRD)-funded project.

The paper "Algorithms for Quad-Double Precision Floating Point Arithmetic" was presented at the ARITH-15 conference. The paper "A Compendium of BBP-Type Formulas for Mathematical Constants" was completed. This paper gives numerous experimentally discovered new mathematical identities. The talk "High Precision Arithmetic and the IEEE Floating-Point Standard" was given at UC Berkeley.

The paper "On the Random Character of Fundamental Constant Expansions" appeared in the journal *Experimental Mathematics*. This paper demonstrates that the age-old question of the randomness of the digits of  $\pi$  (and certain other mathematical constants) can be reduced to a simple and plausible hypothesis from the field of chaotic sequences. This publication attracted international attention from the scientific press:

- LBNL press release: <http://www.lbl.gov/Science-Articles/Archive/pi-random.html>
- DOE Office of Science home page: [http://www.sc.doe.gov/feature\\_articles\\_2001/july/Digits\\_of\\_Pi/Digits%20of%20Pi.htm](http://www.sc.doe.gov/feature_articles_2001/july/Digits_of_Pi/Digits%20of%20Pi.htm)
- *Nature* on-line magazine (Aug. 2, 2001): <http://www.nature.com/nsu/010802/010802-9.html>

- *Science* magazine (Aug. 3, 2001 issue):  
<http://www.sciencemag.org/cgi/content/full/293/5531/793>
- *Science News* (cover story, Sept. 1, 2001 issue):  
<http://www.sciencenews.org/20010901/bob9.asp>
- *Neue Zürcher Zeitung*, (Switzerland, Oct. 31, 2001 issue): <http://www.nzz.ch/2001/10/31/fv/page-article7Q81D.html>

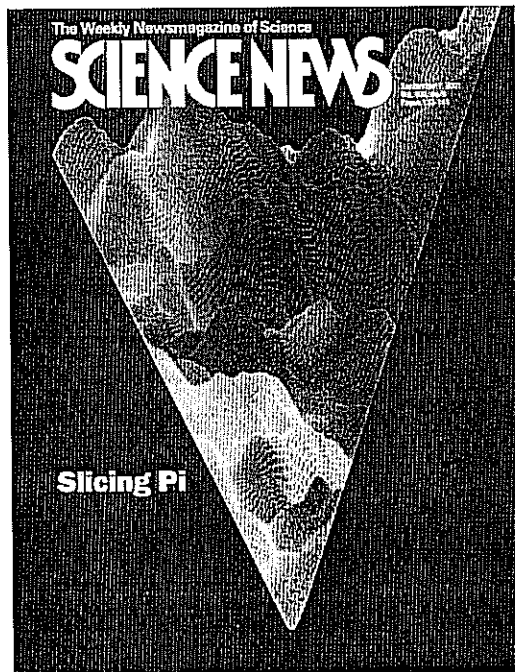


Figure 9: Cover of *Science News* magazine, September 1, 2001 edition.

## Publications

D. Bailey and R. Crandall, "On the Random Character of Fundamental Constant Expansions," *Experimental Mathematics*, LBNL-45583 (June 2001).

Y. Hida, S. Li, and D. Bailey, "Algorithms for Quad-Double Precision Floating Point Arithmetic," published in proceedings of *ARITH-15 Conference*, LBNL-46996 (June 2001).

S. Li, J. Demmel, D. Bailey, G. Henry, Y. Hida, J. Iskandar, W. Kahan, A. Kapur, M. Martin, T. Tung, and D. Yoo, "Design, Implementation and Testing of Extended and Mixed Precision BLAS," submitted to *SIAM Journal of Mathematical Software*, LBNL-45991 (October 2000).

D. Bailey, "A Compendium of BBP-Type Formulas for Mathematical Constants," LBID-2383 (November 2000).

---

## ***Nonlinear Mathematical Models of Phenomena Related to Petroleum, Mining, and Geological Engineering***

---

Principal Investigators: Grigory Barenblatt

Project No.: 01012

## **Project Description**

An overarching motivation for this project is to apply non-linear mathematical models for study of the safety and reliability of proposed waste disposal schemes. More specifically, a set of asymptotic and numerical methods

will be used for solving the equations of damage accumulation in aggressive environments and fissured rocks.

In addition, we will investigate the applicability of methods of mathematical modeling and current technology used in engineering practice to model the development of oil deposits in diatomites. Such oil deposits form a substantial part of national oil reserves, and, in particular, an important part of California's oil reserves (Lost Hills and Belridge containing more than 10 billion barrels of hydrocarbons). Similar rocks form oil collectors in the Northern Sea, and apparently in Northern Siberia. Diatomites are rocks with high porosity (50 to 70%), extremely low permeability (hundreds of times less than solid cement), and low fracture toughness.

### Accomplishments

The injection of carbon dioxide ( $\text{CO}_2$ ) to gas deposits is now widely discussed as being one of the prospective methods of sequencing carbon dioxide from the atmosphere and simultaneously improving gas recovery. A nonlinear mathematical model of such a process was constructed. The solution was obtained by approximate integral methods. The accuracy of the methods was approved by comparison with an exact solution for a special case. The results are presented in a form that allows engineers to estimate the influence of parameters, such as thickness of the layers occupied by carbon dioxide and methane ( $\text{CH}_4$ ), temperature, pressure, etc. The characteristic times of mixing of carbon dioxide and methane, which are of basic practical interest, were obtained for important cases. Up to now only a limited set of special cases was considered; the continuation of this work requires additional analytic investigations and numerical computations.

A second detailed investigation has been performed of non-equilibrium effects in processes of two-phase flows in rocks, such as capillary counter-current imbibition and water-oil displacement. Such processes play an important role in secondary oil recovery and liquid waste storage. A critical analysis of the literature was completed taking into account both Western and Russian studies. Also, analytic and numerical investigation of the process of counter-current capillary imbibition was performed, taking into account the saturation dependence of the relaxation time. The results were compared with recent experimental data, and excellent agreement was obtained. The basic conclusion is that both counter-current imbibition and displacement of non-wetting fluid by a wetting one cannot be calculated using commonly used methods based on the assumption of local equilibrium. The analysis performed allows us to recommend proposed methods for practical use.

Finally, analysis of the development of oil deposits in diatomites performed in our work showed that the current practice of analysis and development of such reservoirs is inappropriate. Its application can lead ultimately to reduced oil recovery and massive destruction of well-bores. A principally new mathematical model for the development of the oil fields in diatomites (such as Lost Hills and Belridge in California) is needed. The general requirements for the construction of such models have been formulated and a program of experimental and numerical investigations has begun.

### Publications

T.W. Patzek, D.B. Silin, S.M. Benson, and G.I. Barenblatt, "On Vertical Diffusion of Gases in a Horizontal Reservoir," LBNL-48859, (submitted to *Transport in Porous Media*).

T.W. Patzek, D. B. Silin, and G.I. Barenblatt, "Impact of Rock Micro- and Macrostructure of the Behavior of Two-Phase Flow," *Proceedings of the 22nd Annual International Energy Agency Workshop and Symposium*, Vienna, Austria, (September 10-12, 2001).

G.I. Barenblatt, T.W. Patzek, and D.B. Silin, "Spontaneous Countercurrent Imbibition: Mathematical Modeling Survey," (in preparation).

D.B. Silin, G.I. Barenblatt, and T.W. Patzek, "Spontaneous Countercurrent Imbibition: Self-Similar Asymptotic Solutions," (in preparation).

---

### Science-Based Subgrid Scale Modeling in Fluid Turbulence

---

Principal Investigators: John Bell, Phillip Colella, Alexandre Chorin, and Nancy Brown

Project No.: 00006

### Project Description

We propose the development of an integrated approach to subgrid scale model development for reacting fluid flows. This project will build on the combined strengths of a number of groups at Berkeley Lab: in the understanding of fundamental analysis of fluid dynamics and chemical reactions; in large-scale simulations of fluid dynamics; and in parallel computing. We expect to develop and validate a number of new models for turbulence, in particular turbulent boundary layers and turbulent reacting

fluid flows. In addition, we will develop the software capability required to support an integrated approach to model development, and will make this capability available to the Department of Energy (DOE) scientific community.

We will use new and more rigorous methodologies for developing subgrid scale models for two key problems in turbulence. For turbulent boundary layers, we will use the incomplete similarity and optimal prediction approaches developed in the Berkeley Lab Mathematics Department. For turbulent combustion modeling, we will use the Piece-Wise Reusable Implementation of Solution Mapping (PRISM) methodology for developing reduced descriptions of chemical reactions and transport. The development and validation of these models will be done using a state-of-the-art computational fluid dynamics capability developed in the National Energy Research Scientific Computing Center (NERSC) Division, based on massively parallel implementations of adaptive mesh refinement methods.

## Accomplishments

The overall goal of this research is to develop models for reacting flows that preserve both the chemical and fluid-dynamical fidelity of the flow. However, as simulation complexity increases, it becomes increasingly difficult to extract detailed quantitative information about the flame from the numerical solution, particularly regarding the details of chemical processes. During the past year, we have focused on developing the tools necessary to understand in detail the predicted chemical processes occurring in a flame. As a model problem for this study we focused on the conversion of fuel-bound nitrogen. Both fossil fuels and renewable biofuels contain traces of nitrogen-bearing chemicals. Biofuels typically release their fuel-bound nitrogen in the form of ammonia ( $\text{NH}_3$ ), which the combustion process may convert to the common pollutant, nitric oxide (NO). The amounts of nitrogen involved, though small, can impact emission controls.

Simulations of nonpremixed, laminar, methane flames were performed to study the conversion of ammonia to nitric oxide. The results show that, as ammonia seeding increases, the conversion efficiency to nitric oxide decreases due to increased activity of reaction paths that produce harmless molecular nitrogen ( $\text{N}_2$ ). These findings are corroborated by laboratory measurements. A chemical model developed by Glarborg, *et al.* was found to provide the best agreement with experiment. That model consists of 65 chemical species and 447 reactions.

We have performed simulations using this model across a broad range of operating conditions using our low Mach number adaptive mesh refinement (AMR) methodology. The ability to perform these simulations efficiently permits a thorough examination of the problem's

parameter space. However, the complexity of the resulting datasets makes analysis of the chemical processes in the flame extremely difficult and increases the desirability of analyzing them automatically. For reacting flows, reaction path analysis (RPA) is an appropriate data reduction technique that is customarily done by hand, but with some refinement it is suitable for automation. RPA essentially removes the spatial aspect to provide a summary of just the chemical reactions occurring in a flow field.

A general tool for reaction path analysis was created and tested on the ammonia-methane flame data. Figure 9 depicts the operation of the RPA tool. The cartoon on the left in the figure represents data from an AMR simulation. In this case, the data consists of two-dimensional fields for 65 chemical species and derived fields for 447 reactions. The RPA tool combines and reduces this data to the reaction path diagram pictured on the right. The diagram shows the several routes by which ammonia is converted to either nitric oxide or molecular nitrogen. Analysis of simulation data using this tool has enabled us to determine the role of different chemical reactions controlling  $\text{NO}_x$  production in non-premixed systems. This new capability provides an important part of the functionality required to understand chemical processes in more complex settings such as turbulent flames.



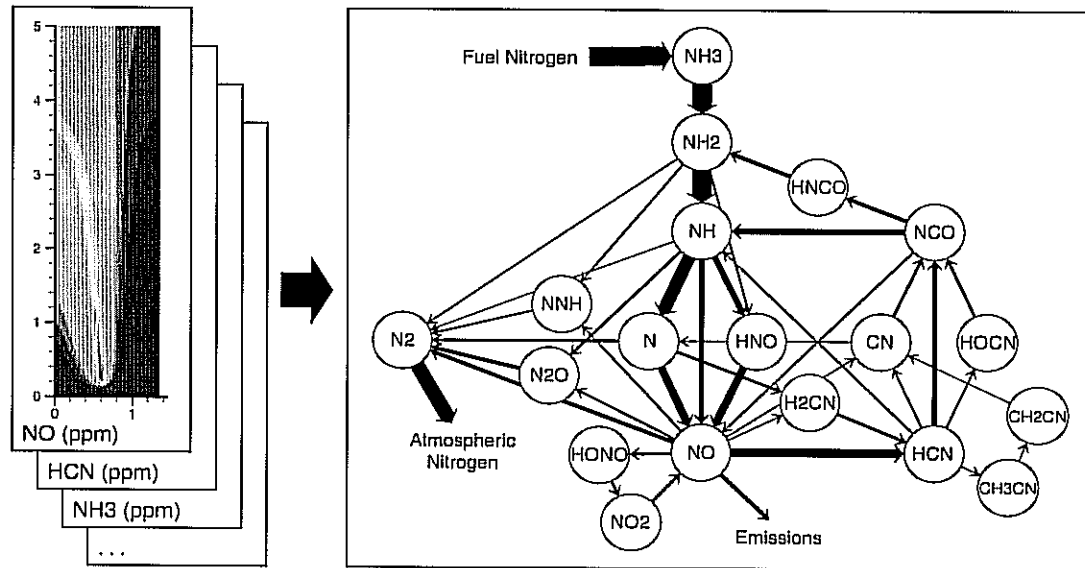


Figure 10: The reaction path analysis post-processing tool reduces the output from a reacting flow simulation to a reaction path diagram. Line widths indicate the amount of nitrogen (mole/second) transferred among chemical species as a result of chemical reaction. The nitrogen in nitric oxide recycles through carbon-bearing species back to the main reaction path where it has an opportunity to bleed away to atmospheric nitrogen.

## Publications

N. Sullivan, A. Jensen, P. Glarborg, M. Day, J. Gracar, J. Bell, C. Pope, and R. Kee, "Ammonia Conversion and  $\text{NO}_x$  Formation in Laminar Coflowing Nonpremixed Methane-Air Flames," submitted to *Combustion and Flame* <http://seesar.lbl.gov/ccse/Publications>.

## Interactive Stereo Electron Microscopy Enhanced with Virtual Reality

Principal Investigators: E. Wes Bethel and S. Jacob Bastacky

Project No.: 01013

## Project Description

An analytical system was developed to take measurements of objects perceived in stereo image pairs obtained from a scanning electron microscope (SEM). Our system

operates by presenting a single stereo view that contains stereo image data obtained from the SEM, along with geometric representations of two types of virtual measurement instruments, a "protractor" and a "caliper". The measurements obtained from this system are an integral part of a medical study evaluating surfactant, a liquid coating the inner surface of the lung that makes possible the process of breathing.

Measurements of the curvature and contact angle of submicron diameter droplets of a fluorocarbon deposited on the surface of airways are performed in order to determine surface tension of the air/liquid interface. This approach has been extended to a microscopic level from the techniques of traditional surface science by measuring submicrometer rather than millimeter diameter droplets, as well as the lengths and curvature of cilia responsible for movement of the surfactant, the airway's protective liquid blanket. An earlier implementation of this approach for taking angle measurements from objects perceived in stereo image pairs using a virtual protractor is extended to include distance measurements and to use a unified view model.

The system is built around a unified view model that is derived from microscope-specific parameters, such as focal length, visible area, and magnification. The unified view model ensures that the underlying view models and

resultant binocular parallax cues are consistent between synthetic and acquired imagery. When the view models are consistent, it is possible to take measurements of features that are not constrained to lie within the projection plane.

The system is first calibrated using non-clinical data of known size and resolution. Using the SEM, stereo image pairs of grids and spheres of known resolution are created to calibrate the measurement system. After calibration, the system is used to take distance and angle measurements of clinical specimens.

## Accomplishments

The unified view model creates a framework whereby binocular parallax cues are consistent between two types of views. One type of view is the binocular stereo view frustum needed to render views of geometric models of virtual sensors. The sensors are used to obtain distance and angular measurements. The other binocular stereo view of the specimen obtained by either eucentric tilting or view-plane translation within the instrument.

A generalized view frustum for the SEM instrument can be approximated by  $\alpha = \arctan(1/2x)$ , where  $x$  is the magnification scale factor. This generalization must be tuned to each particular instrument, and consists of several instrument-specific parameters. These include a focal length  $f$ , a measure of viewable area  $V$ , a magnification factor  $x$ , and a nominal imaging resolution  $l$ . These parameters are combined to create a binocular view

frustum used for rendering stereo views of sensor geometry. When the rendering frustum is the same as the imaging frustum, a single scale factor can be applied to sensor geometry coordinates in order to obtain distance measurements, thereby avoiding unnecessarily complex coordinate system conversions.

Prior to using the caliper sensor on actual medical data, we first performed a calibration step to verify that the view frustum models were indeed correct. We obtained stereo images of a 2000 etched copper mesh at 1000x and 5000x. From these images, we were able to obtain measurements using a digital caliper that exhibit an error of less than approximately 0.5%.

We obtained stereo images of a fractured droplet of fluorocarbon deposited on the surface of the trachea. The height of the relatively flat drop is measured with the virtual caliper to be approximately 15.5 microns high. The width of the droplet is considered the average of the major and minor axes of the roughly elliptical drop, whose border can be barely distinguished in this micrograph. The width of this droplet is approximately 256 microns. The ratio of height to diameter is inversely proportional to surface tension of the surface upon which the droplet rests. In this case, surface tension of the surface of the trachea is higher than the surface tension of the fluorocarbon (16 dynes/centimeter) so the drop is pulled flat. We expect surface tension to be different at other places in the lung, and are beginning to use the software to measure the variation in surface tension as a function of position along the airway in the mammalian lung.

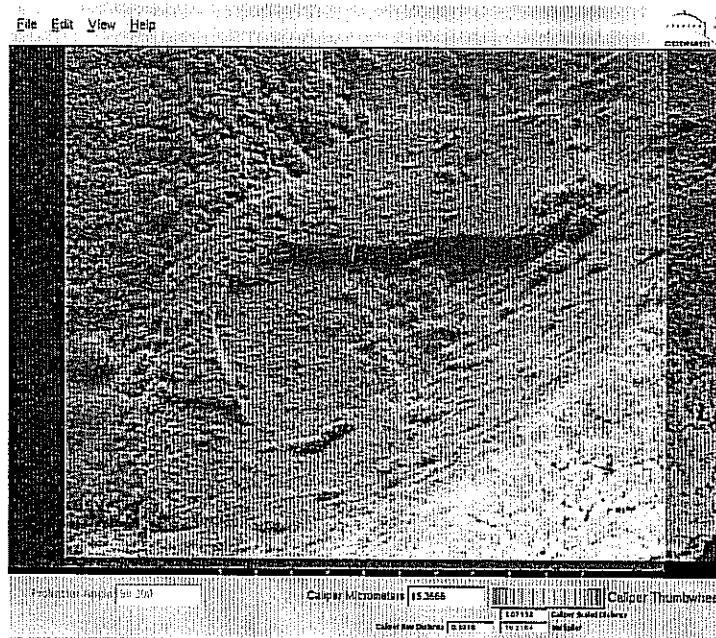


Figure 11: A fractured droplet of fluorocarbon deposited on the surface of the trachea as described in the text.

## Publications

E.W. Bethel and S.J. Bastacky, "Interactive Stereo Electron Microscopy Enhanced with Virtual Reality," *Proceedings of SPIE/IS&T (International Society of Optical Engineering, Society for Imaging Science and Technology) Photonics West Symposium on Imaging Science and Technology, The Engineering Reality of Virtual Reality*, San Jose, California (January 2002).

---

## Computational Methods for Electronic Structure Codes in Materials Science: Ground-State and Excited-State Properties

---

Principal Investigators: Andrew Canning and Steven Louie

Collaborator: Lin-Wang Wang

Project No.: 00007

## Project Description

A fundamental understanding of the properties of a material requires the knowledge of its electronic structure as determined by the valence electrons. In particular, as electronic components move towards the nanometer scale, quantum effects become increasingly important and can only be predicted and understood from accurate calculations of the electronic structure.

This project aims to develop state-of-the-art electronic structure methods and parallel codes and make them available to the materials science community and in particular the Department of Energy (DOE) community and National Energy Research Scientific Computing Center (NERSC) users. These codes will perform ground-state as well as excited-state electronic structure calculations. The ground-state calculations are based on density functional theory (DFT) using pseudopotentials to represent the atomic core regions and the local density approximation (LDA) or the generalized gradient approximation (GGA) for the electron-electron interaction. The wavefunctions for the electrons are expanded in plane-waves. Codes for electronically excited states will also be developed using a first-principles approach developed by Louie and collaborators based on the GW approach (The "G" in GW refers to the dressed Green's function of the electron and the

"W" is the screened coulomb interaction.) In the case of optical properties where an electron-hole pair (exciton) is formed, the interaction between the excited electron and the hole is included through the two-particle Green's function by solving the Bethe-Salpeter (BS) equation. The excited-state codes require the ground-state wavefunctions and energies as inputs, so the complete set of codes is required for an excited-state calculation. Development of these codes and methods will be applications-driven from physical problems relevant to the Materials Sciences (MSD) and Advanced Light Source (ALS) divisions at Berkeley Lab. These types of electronic structure calculations are very demanding of computer power and are a natural match for the large parallel computers at NERSC.

## Accomplishments

### Ground-State Methods and Code

In the DFT code PARATEC (PARAllel Total Energy Code) a Grassman conjugate gradient method for metals has been introduced into the code for the minimization of the electronic degrees of freedom, which has advantages over the unconstrained energy functional previously used in the code, for certain types of system. This approach gives much better convergence for metals that have traditionally been very difficult to study with standard techniques. Many of the standard methods developed for systems with a gap (i.e. insulators and semiconductors) give very poor or no convergence when applied to metals. Many other improvements have also been made to PARATEC including the ability to accept different formats for pseudopotentials that model the core region of the atoms. In particular, the program accepts different formats of inputs for pseudopotentials from the most heavily used, public domain pseudopotential codes. The communication structures in the code have also been changed to run more efficiently on the new IBM SP at NERSC.

A web site has been constructed for the code ([www.nersc.gov/projects/paratec/](http://www.nersc.gov/projects/paratec/)) that contains information on the code, a user's guide, a list of updates in the new versions of the code, a list of credits of contributors to the code and a registration form for obtaining the code. At Berkeley Lab in addition to Steven Louie's (MSD) group who is using the code, Marvin Cohen's (MSD) group is using the code to study the upper limits of the strength of metals in collaboration with John W. Morris's group (MSD). Daryl Chrzan's group (MSD) is looking at dislocation cores using the code.

### Excited-State Methods and Codes

Preliminary parallel versions of the GW and exciton codes have now been developed that are used by other groups who are part of the DOE computational materials science network (CMSN) project on excited states. (The DOE CMSN project on "Excited-State Electronic Structure and

Response Functions" is coordinated by Steven Louie at Berkeley Lab and John Rehr at the University of Washington.) These codes are optimized for large-scale parallel machines such as those at NERSC and take as their input the ground-state wave functions calculated by PARATEC. One of the central computational burdens in the GW+BS method is the calculation of large numbers of matrix elements, each matrix element being the Fourier expansion coefficient of various combinations of excited electronic states. Conventionally, the matrix elements are computed directly in Fourier space as a convolution and the calculation of the needed matrix elements scales as  $O(n^2)$  where  $n$  is the number of Fourier coefficients in the expansion. This can be reformulated in real space using fast Fourier methods, which leads to a calculation scaling as  $O(n)$  with a larger pre-factor. This leads to a dramatic speedup for large systems and in the case of a large benzene simulation gave a speedup on the order of thirty. The fast Fourier methods have been implemented in both the GW and exciton codes.

The calculation of forces for excited-state configurations is also being developed. In many cases once the electrons are excited above their native state (e.g. when light is absorbed) their new configuration creates forces on the atoms, which cause the atomic structure to distort thereby changing the electronic properties as well. An example of this is photoluminescence, whereby a material absorbs light at one frequency, rearranges its structure, and then emits light (de-excites) at a different frequency. We have developed a new formalism where we use the excited-state wave functions to compute the forces on the atoms for excited electronic states, allowing us to optimize the atomic positions. This method allows the relaxation of excited electronic states with an effort that is no more than the actual exciton calculation.

The GW+BS codes have been used in large-scale calculations such as the electronic spectrum of benzene, a prototypical organic molecule with an aromatic structure. Aside from the intrinsic interest of benzene, we have also been comparing the GW+BS method to other excited-state methods such as time-dependent, density functional theory. Our preliminary results are promising in that the energies of the low-lying transition states are well described when compared to experimental results. These calculations are very demanding, as the molecule is relatively large and the calculation was only possible using the new real-space approach.

## Publications

G.M. Rignanese, S.G. Louie, and X. Blase, "Quasiparticle Band Structure of  $C_2H_4$  Adsorbed on the Si(001)-(2x1) Surface," *Physical Review Letters*, **86**, 2110 (March 2001).

D. Raczkowski, A. Canning, and L-W Wang, "Thomas-Fermi Charge Mixing for Obtaining Self-Consistency in

Density Functional Calculations," *Physical Review B*, **64**, 121101 (September 2001).

S. Ismail-Beigi, E.K. Chang, and S.G. Louie, "Coupling of Nonlocal Potentials to Electromagnetic Fields," *Physical Review Letters* **87**, 087402 (August 2001).

J.R. Chelikowsky, S. Ogut, S. Ismail-Beigi, and S.G. Louie, "Comparison of Time Dependent Local Density Approximation and Bethe-Salpeter Methods for the Optical Properties of Covalent and Ionic Systems," abstract published in *Proceedings of American Physical Society March Meeting*, Seattle, Washington (March 2001).

---

## Atomic-Scale Modeling of Materials Chemistry

---

Principal Investigators: Niels Jensen

Project No.: 01014

## Project Description

Shock physics, materials chemistry, and non-equilibrium statistical mechanics are all integrated components in studying detonation of chemically active atomic-scale materials. The purpose of the proposed work is to model chemically active materials at the atomic scale and to answer fundamental questions such as: what reaction pathways are likely to exist in 'n'-component reactions; shock-front speed and width; the detailed relationship between shock front and reaction front during the process; and the threshold values for sustained reaction propagation in different materials and structures with and without disorder. Our atomic-scale results can be compared directly to results of continuum models describing chemical reactions at the macroscopic level.

This topic has wide interest in many scientific and industrial contexts, such as ion implantation of wafers where chemical reactions occur due to the deposited energy. We will apply large-scale molecular dynamics simulations of material composites consisting of several different types of atoms, all interacting with Brenner-Tersoff type potentials. The model material can be initiated in a number of different metastable states, after which a perturbation can provoke a sustained (or unsustained) chemical reaction of local structural atomic reorganization. This type of simulation is ideally suited for large-scale molecular dynamics, since we may easily require simulations of tens of millions of atoms, while the simulation time is limited by the fast timescales of active chemical reactions.

## Accomplishments

The project has evolved in two directions; one is to extend the investigations of the materials chemistry of two-dimensional atomic-scale behavior, another is to extend the code to simulate three-dimensional materials.

Two-dimensional reactive materials have been investigated with respect to critical activation of materials reaction as a function of large imperfections (density fluctuations) in the material. For example, we have determined how both voids and stiff regions in a reactive material serve as nucleation for the onset of reactions, thereby decreasing the critical initiation parameters for reaction when almost any density anomaly is present in the material. Extensive simulations have been conducted to map the dependencies of the initiation of the reaction upon the size, shape, and type of density anomalies. This work has also led to an investigation of how the initiation depends on surface roughness of the material.

We have conducted extensive simulations to determine how stable reaction fronts propagate in small finite size systems. It has previously been established in the literature that for given chemical bond energies a critical width of material is necessary to sustain a steady-state propagation of shock/chemistry fronts. We have determined, for the commonly used Brenner-type bond order potential, how this critical width depends on the energy release of each two reacting atomic dimers.

Preliminary simulations of three-dimensional reactive materials have been conducted to determine the critical width (radius) beyond which reaction fronts can be sustained and we believe we have found the critical radius for one set of parameters.

All work is done in collaboration with scientists at Los Alamos National Laboratory. We are currently writing a manuscript detailing and summarizing the above-sketches results as well as the results on dirty and disordered materials obtained during FY 2001.

## Publications

D. Heim, N. Gronbech-Jensen, T. Germann, B.L. Holian, and P. Lomdahl, "Initiation and Propagation of Reaction Fronts in Chemically Active Materials," in preparation for *Physical Review B*.

---

## ***Suitability of Alternative Architectures for Scientific Computing in the NERSC-5 Time Frame***

---

Principal Investigators: Xiaoye Li, Leonid Oliker, and Katherine Yelick

Project No.: 01015

## Project Description

As we look beyond NERSC-4, important questions arise regarding how key scientific applications will perform on future generation supercomputers. A new class of microprocessors with imbedded memory is emerging from several leading academic and industrial institutions. We plan to evaluate the feasibility of using large-scale multiprocessor systems based on this technology for key scientific computations at the National Energy Research Scientific Computing Center (NERSC).

To accomplish this, we will evaluate how key computational codes of NERSC and Department of Energy (DOE) will perform on future generation architectures. These include climate modeling, combustion, material science and quantum chemistry. We will also incorporate the novel architectural research being developed at University of California (UC), Berkeley. In addition to porting codes onto prototype systems, we will use simulation and analytic modeling to predict the expected performance of these applications on large-scale systems. Based on detailed simulations and executions, we will modify the algorithms to better fit the architecture and improve the compiler and performance tools. We will also examine the network requirements of this unique cluster, where the balance of system characteristics is significantly different from conventional architectures.

## Accomplishments

We have been working closely with the VIRAM software group at UC Berkeley, led by Professor Yelick. The VIRAM is an example of a processor-in-memory (PIM) or intelligent random access memory (IRAM) design. It combines mixed logic and dynamic RAM (DRAM) with a vector instruction set, allowing the compiler to explicitly express fine-grained data parallelism. The VIRAM group has developed several simulation tools including a functional instruction set architecture (ISA) simulator; a debugging version of the ISA simulator; and a cycle level performance simulator for the vector unit and memory

system. The project has also ported a vectorizing compiler for C/C++, based on Cray's commercial compiler PDGCS.

Last year we studied the performance of FFT, DCT, and some dense linear algebra kernels. Recently, we have focused on using the VIRAM performance simulator to understand the performance of several memory-intensive kernels that are common in scientific computing. These include: transitive closure, GUPS (Giga-Updates-Per-Seconds), sparse conjugate gradient, histogram, and unstructured mesh adaptation. We studied various characteristics of these kernels, such as memory access patterns, ratio of computation-to-memory operations, and degree of irregularity in their basic controls. All of these benchmarks are memory-intensive in the following sense: the number of arithmetic or logical operations per step of

the algorithm is never more than the number of memory operations.

For all these kernels, we compared the VIRAM simulation results with the results obtained from many commercially significant architectures, including Sun UltraSPARC, MIPS R10000, Pentium III, Pentium 4, and Compaq Alpha EV6. We found that VIRAM is significantly faster than the cache-based machines for problems that are limited only by DRAM bandwidth and latency. Since VIRAM achieves its high performance through parallelism rather than a fast clock rate, the advantages are even larger if one is interested in building an energy-efficient system for scientific computation: compare hundreds of Watts for a Pentium-based processing node with two Watts for VIRAM.

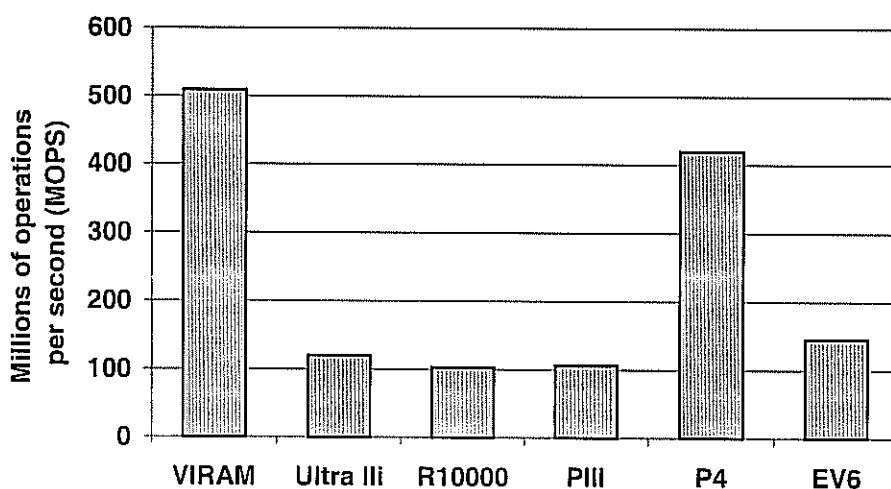


Figure 12: Performance of the unstructured mesh adaptation in millions of operations per second (MOPS), on various architectures.

## Publications

B. Gaeke, P. Husbands, X. Li, L. Oliker, K. Yelick, and R. Biswas, "Memory-Intensive Benchmarks: IRAM vs. Cache-Based Machines," *International Parallel and Distributed Processing Symposium* (April 15-19, 2002) (in press) LBNL-48979.

---

## Berkeley Lab Distribution (BLD): Software for Scalable Linux Clusters

---

Principal Investigators: Paul Hargrove, William Saphir, and Robert Lucas

Project No.: 99013

## Project Description

An exciting development in scientific computing over the past few years has been the demonstration that clusters of

personal computers (PCs) can achieve the performance of supercomputers for many problems of interest to the Department of Energy (DOE). Such clusters promise to greatly increase the impact of computation on science. Despite their promise, we believe that clusters are destined for only a minor role unless two critical barriers are overcome:

- Most existing and planned system software for clusters is not scalable, making infeasible large clusters that can be applied to DOE mission-critical applications.
- Cluster software is ad-hoc, poorly integrated, lacking in functionality, and difficult to use, making the setup and management of clusters very labor intensive, and limiting their flexibility.

We are designing, implementing, and integrating scalable system software and architectures for clusters. This software will form the basis of the "Berkeley Lab Distribution," or BLD, a complete package of plug-and-play software for configuring, managing, and using Linux clusters.

## Accomplishments

This project began by focusing on understanding cluster requirements, developing basic infrastructure for the cluster, and building collaborations with other laboratories in order to form the basis for coordinated multi-lab system software development. This included releasing trial distributions, which enabled us to collect early feedback from potential users of BLD systems, and to perform a detailed study of cluster software deployment.

We have evaluated dozens of tools to be used as software components in BLD. The results of these evaluations have been used to elicit requirements for future software development projects, and allowed us to participate in the open source community. We have integrated some of these tools into our existing cluster system. Tools evaluated include Chiba City, SCMS, Rexec, SGI ACE, Globus tools, FAI, SGI PCP, and PVFS.

This project has developed software called BCM (Berkeley Configuration Manager). BCM significantly simplifies the tasks of installing and maintaining file systems for the compute nodes of a cluster while using a minimum amount of storage on the file server. One significant feature of BCM is its natural support for heterogeneous clusters. BCM also simplifies the changes needed to add, remove, replace, or reconfigure compute nodes. BCM is designed from the beginning to be scalable. While the present implementation only supports diskless compute nodes, the software has been designed in a modular format to easily accommodate components that support diskfull compute nodes. BCM has been deployed

to maintain our own development clusters. Several other groups at Berkeley Lab have deployed BCM or are considering doing so. BCM was used to configure the cluster used to win the High-Performance Bandwidth Challenge at the SC2001 Conference.

Additional new tools were developed for process startup and accounting on BLD clusters. Tmrsh is a tool for process startup that addresses some important deficiencies in previous tools—starting jobs twice as fast as other tools, and providing for better fault tolerance while the cluster is operating. Acctpbs is a tool for analyzing cluster accounting data. Acctpbs allows the administrator to generate specialized reports to analyze the important aspects of cluster system utilization including total central processing unit (CPU) time usage, total system utilization (average or interval), and other important metrics necessary in a production-quality computing environment.

We have also maintained a collection of benchmark results for processors used in Linux clusters. These results have become a valuable and highly visible resource for the community. These may be seen at:  
<http://www.nersc.gov/research/ftg/pcp/performance.html>.

This project has lead directly to follow-on funding from DOE in the form of participation in the Scalable Systems Software Integrated Software Infrastructure Center. Funded as part of DOE's Scientific Discovery through Advanced Computing (SciDAC) initiative, this is a multi-lab effort to develop scalable implementations of systems software for clusters, and to specify interfaces that ensure interoperability of systems software components.

---

## ***Electron Collision Processes above the Ionization Threshold***

---

Principal Investigators: C. William McCurdy and Thomas Rescigno

Project No.: 99011

## **Project Description**

The computational prediction of the electronic structure of atoms and molecules has become a practical task on modern computers, even for very large systems. By contrast, sophisticated calculations on electronic collisions have been limited in terms of the complexity of the targets that can be handled and the types of processes that can be studied. Collision processes are, nevertheless, central to

the problems of interest to the Department of Energy (DOE), playing a key role in such diverse areas as fusion plasmas, plasma etching and deposition, and waste remediation. The intermediate energy region, extending from the ionization threshold to a few hundred eV, presents the greatest challenge for *ab initio* theory, since the infinity of energetically accessible final states precludes one from writing down a wave function that describes all possible scattering events, and is simply a reflection of the fact that ionization persists as one of the fundamentally unsolved problems of atomic collision theory.

A practical route to solving the intermediate energy electron collision problem cannot rely on close-coupling approaches, but rather must be based on formalisms that allow the wave function itself to be computed without recourse to the explicit asymptotic form, and on methods that extract cross section information from the wave function without knowing its detailed asymptotic form. The purpose of this project is to develop such an approach for both atoms and molecules. The approach will build on the algebraic variational formalism we have previously developed to study electron-molecule scattering. The approach will be extended to include the scattered-wave, flux operator formalism, which we have developed, complex optical potential interactions, and a variety of techniques based on analyticity. For atomic ionization problems, we will carry out direct solutions of the Schrödinger equation on a (complex) numerical grid and use the projected flux operator formalism to extract total and differential ionization cross sections.

## Accomplishments

Exterior complex scaling is the formal mathematical tool that we use as the basis for our computational approach to collisional ionization problems. That approach, which has produced the first essentially exact treatment of electron-impact ionization of atomic hydrogen, simplifies the computation of the complete system wave function by obviating the need for a detailed specification of asymptotic boundary conditions. During this past year, we have significantly refined and improved on the basic methodology. In our original implementation, we obtained the ionization cross sections by evaluating the quantum mechanical flux through a finite hypersphere that bounded the region where we knew the wave function and then extrapolated the result to infinite volume. That procedure requires very large grids and has inherent numerical errors. We have since developed a new procedure, based on formal distorted-wave rearrangement theory, for computing the ionization amplitudes directly from the computed wave function. The new procedure is far more efficient, can be used with smaller grids and allows us to compute differential ionization cross sections over the entire range of energy sharing. Using this method, we

have produced and published definitive triply differential ionization cross sections for atomic hydrogen, which validate our earlier results and extend their range. In particular, we have now been able to probe the threshold region for the first time with a fully quantum mechanical treatment, as previous investigations of this region were all based on classical and semi-classical theory.

We have also extended the formalism to treat rearrangement scattering. This extension will allow us to study ionization by positron impact, which is more difficult to treat than the electron-impact case because it is complicated by the possibility of positronium formation competing with breakup. We have tested our ideas by constructing simple two-dimensional models of positron-hydrogen atom scattering and shown that we can devise stable methods for computing all cross sections of interest.

Finally, we have begun to address the problem of electron impact ionization of multi-electron targets. Our original method explicitly solves the time-independent Schrödinger equation on a numerical grid, which results in large systems of complex linear equations that become prohibitively difficult to solve in more than two dimensions. We have therefore developed a time-dependent formulation of exterior complex scaling that enables us to rigorously compute the system wave function by propagating a wave packet on an exterior-scaled grid and accumulate its Fourier transform during the course of propagation. This approach does not involve the solution of large linear systems and hence scales more favorably with particle number than does the time-independent method. We will use the time-dependent approach to study electron-helium ionization.



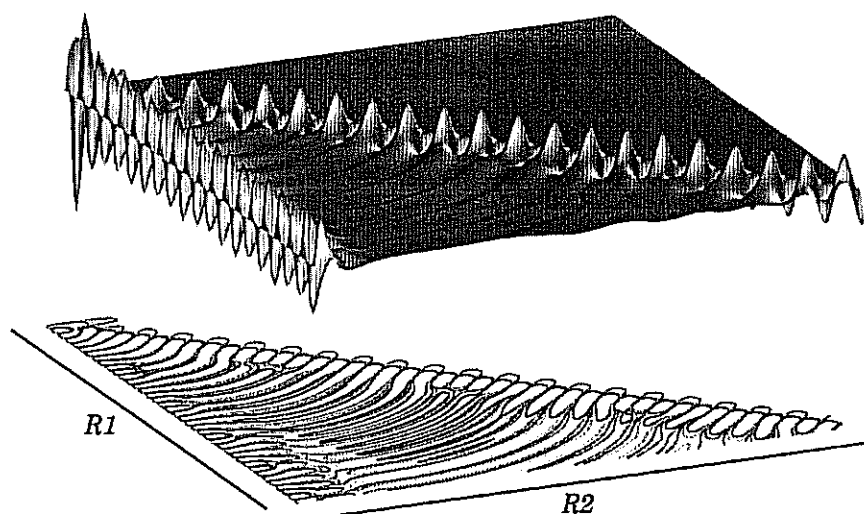


Figure 13: Wave function for a model two-dimensional simulation of positron impact on atomic hydrogen. A 20 eV positron is incident along the  $R1$  axis. The radial waves in the sector  $R1 > R2$  correspond to ionization, leaving a bare proton plus a free electron and a free positron which carries off most of the energy. The concentration of density along the  $R1 = R2$  axis corresponds to rearrangement events that produce bound positronium.

## Publications

C.W. McCurdy, D.A. Horner, and T.N. Rescigno, "Practical Calculation of Amplitudes for Electron-Impact Ionization," *Physical Review A* **63**, 022711 (2001).

M. Baertschy, T.N. Rescigno, W.A. Isaacs, X. Lee, and C.W. McCurdy, "Electron Impact Ionization of Atomic Hydrogen," *Physical Review A* **63** (2001).

W.A. Isaacs, M. Baertschy, C.W. McCurdy, and T.N. Rescigno, "Doubly Differential Cross Sections for the Electron Impact Ionization of Hydrogen," *Physical Review A* **63**, 030704 (2001).

M. Baertschy, T.N. Rescigno, C.W. McCurdy, J. Colgan, and M.S. Pindzola, "Ejected-Energy Differential Cross Sections for the Near Threshold Electron-Impact Ionization of Hydrogen," *Physical Review A* **64**, 022709 (2001).

T.N. Rescigno, W.A. Isaacs, A.E. Orel, H.D. Meyer, and C.W. McCurdy, "Theoretical Study of Resonant Vibrational Excitation of  $\text{CO}_2$  by Electron Impact," submitted to *Physical Review A*.

## Second-Order Methods for Solid-Fluid Shock Coupling with Application to Martian Meteorites

Principal Investigators: Gregory Miller

Project No.: 01016

## Project Description

We are developing new numerical methods to model impact problems that include multiple materials separated by complex time-dependent boundaries. Our goal is an accurate, robust solver that can be used for very large three-dimensional problems by exploiting parallelism and adaptive mesh refinement.

We will apply our new methods to impact problems on planetary surfaces. The specific scientific application we are concerned with is the mechanism by which meteorites have been sent from Mars to the Earth, presumably by

impact, but without showing evidence of having experienced large shock pressures. Two qualitative models have been suggested. In one, jets emitted in an impact propel rocks from the surface—like a fire hose sweeping pebbles off pavement. In the other, an interaction between the compressive shock and zero-pressure surface boundary condition is imagined to provide large accelerations by small pressures. We plan to model realistic scenarios with sufficient fidelity that the feasibility of these proposed mechanisms may be assessed.

The approach we adopt consists of three major components. First, we are developing a new numerical framework for solving multiphase dynamics on Eulerian grids, with adaptivity, and in parallel.

The second step consists of constructing new hyperelastic equations of state and constitutive models. This is necessary to accurately model the response of rocks and ice to high velocity impact. Two key concerns are phase change (melting and vaporization), and fracture (breakup of a solid into a rubble with no strength in tension). Third, there are adjustments to the numerical method that may improve its robustness and accuracy. This involves substantially new algorithm development.

## Accomplishments

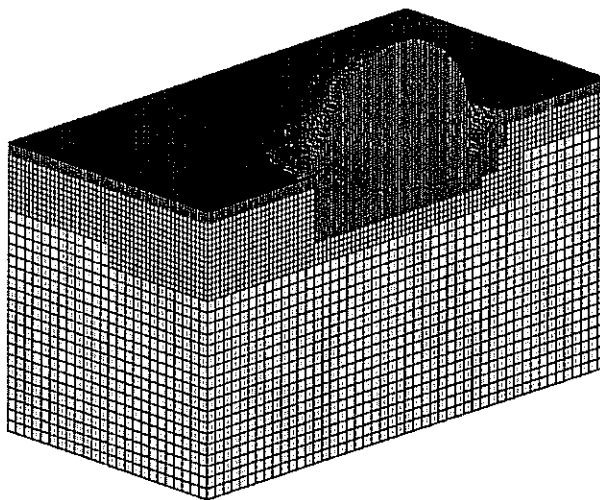
We have developed an implementation of an Eulerian multimaterial shock capturing code in three dimensions. This code uses the Chombo toolkit (<http://seesar.lbl.gov/anag/chombo/>) to exploit parallelism and to implement adaptive mesh refinement.

Our work includes several innovations. First, we developed a new computation method for solid mechanics in an Eulerian framework. This provides a solid mechanics solver that is compatible with state-of-the-art

fluid dynamics solvers, and may easily be applied within frameworks for block-structured adaptivity and parallelism. In the Eulerian framework, the equations of solid mechanics are a system of partial differential equations with constraints. We developed a new approach to solving these constrained systems that will have applicability in other areas, including magnetohydrodynamics.

We have also developed a new approach to modeling multimaterial problems on structured Eulerian grids. We associate with each material a volume fraction, and solve explicit equations for volume fraction advection, which allows the domains to grow, merge, or tear dynamically. In each material domain, the appropriate physics—solid mechanics or fluid dynamics—are solved with high-order shock capturing methods. At boundaries, where a given Eulerian computational cell may contain two or more materials, we have extended the so-called “Cartesian grid” strategy to provide stable and accurate solutions that respect the underlying conservation laws. The order of accuracy of the overall method is lower at boundaries than elsewhere, and we are actively working on solutions to improve this behavior.

With this new code we have been able to model simple impact problems, as shown in the attached figure. This figure shows a snapshot in time of an initially spherical impactor having penetrated the surface of the target material. The initial trajectory was oblique at 45 degrees and 10 kilometers per second velocity. The materials are rock-like elastic-plastic solids. An impact jet appears to be forming at the leading edge of the impactor (left). No evidence for ejection of unshocked surface rocks by interaction of the shock with surface boundary conditions is seen yet.



*Figure 14: The impact of a sphere against a plate at 45 degrees and 10 kilometers per second. Shading corresponds to pressure from 0 (light) to 1.1 million atmospheres (dark).*

## Publications

G.H. Miller and P. Colella, "A High-Order Eulerian Godunov Method for Elastic-Plastic Flow in Solids," *Journal of Computational Physics*, **167**, 131-176 (2001).

G.H. Miller and P. Colella, "A Conservative Three-Dimensional Eulerian Method for Coupled Fluid-Solid Shock Capturing," in preparation for submission to *Journal of Computational Physics*.

---

## Adaptive File Replication and Coordinated Transfer for Data-Intensive Grid Applications

---

Principal Investigators: Ekow Otoo

Project No.: 01017

## Project Description

The objective of this work is to develop adaptive distributed algorithms that coordinate and compute optimal strategies for accessing required subsets from very large datasets on a data-grid. The envisioned environment is a high-speed network that interconnects hierarchical data storage systems, distributed disk caches and computational nodes. The datasets are maintained as collections of files and are distributed over some specialized nodes/sites that have tertiary storage. Computations make requests for files that are subsets of the datasets. The goal then is to develop an optimal strategy for caching and replicating files in distributed caches so that requests are processed efficiently over the network.

Related problems in this area are known to be NP-hard (an optimization problem on a particular complexity class of decision problems referred to as nondeterministic polynomial time algorithms) and hence our goal would be to strive for good heuristic algorithms. The algorithms to be developed would exploit knowledge of the network bandwidth, statistics of data access pattern, and information from meta-data and replica catalogues, to generate an optimal access plan for any applications' data requests. Based on the knowledge of availability of disk cache space, access pattern, and the query classes, the algorithms determine policies for retaining and purging file replicas from the disk caches. The results of this research are applicable to other research projects in scientific computing that are ongoing at Department of Energy (DOE) and other

similar agencies. Examples of these are: Particle Physics Data Grid (PPDG), Earth Science Data Grid (ESDG) and the National Aeronautics and Space Administration (NASA)'s Earth Observation System Data and Information System (EOSDIS).

## Accomplishments

Our solution approach so far consists of decomposing the problem into three special cases:

- File accesses from a Hierarchical Resource Manager (HRM) on the grid,
- File accesses from a Disk Resource Manager (DRM) that also makes remote file accesses to an HRM,
- File accesses from a DRM that also makes remote file accesses to other DRMs and HRMs.

We have developed a cost model for the first case, where the main result derived states that if  $C_{(HSS)}(i)$  is the cost of accessing a file  $i$  of size  $S_i$  from a Hierarchical Storage System then an optimal replacement policy evaluated at time  $t_0$ , calls for evicting from the cache, the file  $i$  that has the minimum utility function  $U_i(t)$  where  $U_i(t)$  is defined as:

$$U_i(t) = (K_i(t_0) / (t_0 - t_{-K})) * (C_{(HSS)}(i) / S_i).$$

$K_i$  is the count of the last  $K$  references and  $(t_{-K})$  is the time that the  $K$ th backward reference was made. The above result has been independently derived using different cost models. The performance metric optimized here is the average cost per reference in accessing a file over the data grid. A comparison of our replacement policy with other popular methods such as Least Recently Used (LRU), Least Frequently Used (LFU), Greedy-Dual Size (GDS), shows that this is indeed the case. We note here also that using other performance metrics such as hit ratio or byte-hit ratio does not necessarily arrive at the same conclusion. The replacement policy based on the above utility function is used in a discrete event simulation model of file accesses over the grid to a Hierarchical Storage System. The figure below depicts some of the essential components, entities, and event messages in our simulation model. The model is currently set up to investigate various options of scheduling file transfer tasks with the optimal replacement policy derived for an HRM's cache, to determine the most effective way of coordinating file transfer requests.

The model shown represents the activities of a single site. Our work replicates the model over multiple sites (some of which need not have a tape storage resource) where the sites are configured into a specific distributed network topology. In particular, we focus our study on the multi-tier network configuration of storage resources to address some of the specific operational requirements of High Energy

Physics experiments of the Particle Physics Data Grid (PPDG). The model of the figure shown below is implemented in C++Sim. A port of the code to Java will be

completed before using the code for simulating a multi-site data access model. This will eventually be used to meet the goals of our proposal.

Queuing Model of a Storage Resource Manager

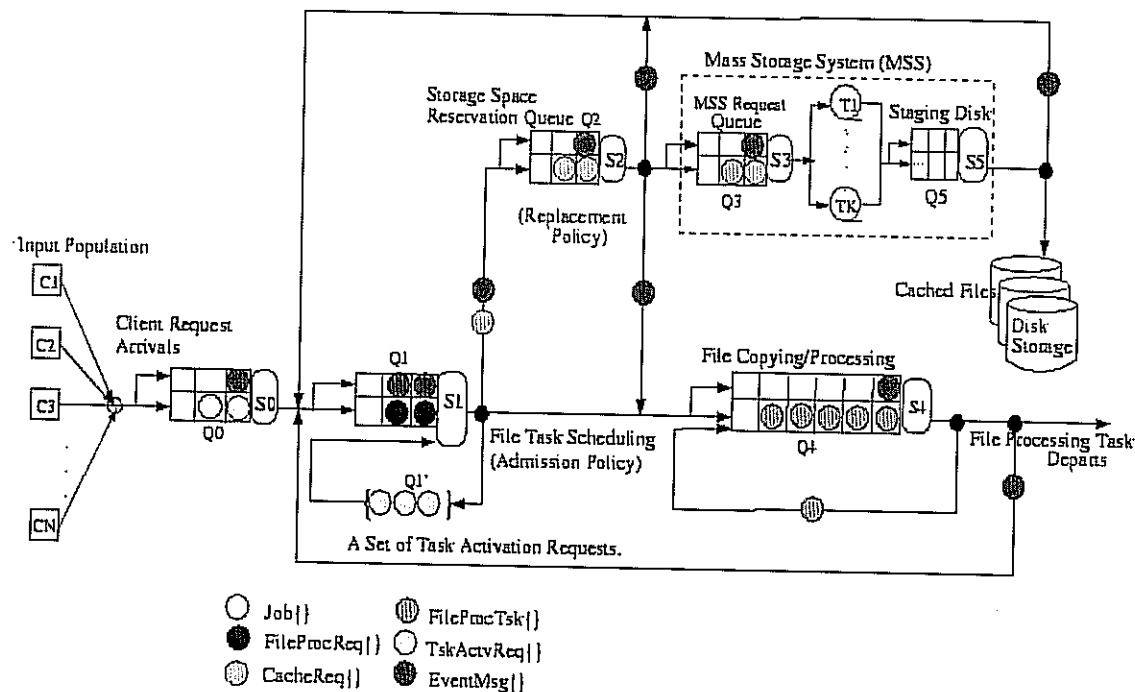


Figure 15: Discrete event simulation model of a Storage Resource Manager

## Publications

E. Otoo, F. Olken, and A. Shoshani, "Efficiently Computing Disk Cache Replacement Policies in Storage Resource Management," presented at GriPhyN (Grid Physic Network) meeting, (October 15, 2001).

E. Otoo, F. Olken, and A. Shoshani, "Efficiently Computing Disk Cache Replacement Policies in Storage Resource Management," (draft).

E. Otoo, D. Guo, and A. Shoshani, "A Discrete Event Simulation Model of a Storage Resource Manager," (in preparation).

## Feature-Based Knowledge Discovery from Multidimensional Data Set

Principal Investigators: Bahram Parvin and Qing Yang

Project No.: 99012

## Project Description

The current generation of scientific instruments/observational platforms generates a massive amount of spatio-temporal images. These images may represent different geophysical fields (e.g., sea surface temperature, wind stress, precipitation, ocean color) at

different spatial and temporal resolutions; physiological responses from specific subcellular compartments of organism to living cells; motions of bacteria in growth media; evolution of a flame front in a combustion engine; or images that are generated from a particular simulation model. The difficulty in comprehending such a vast set of observed or simulated data goes beyond simple visualization and animation of multidimensional data streams. Novel computational techniques must be developed to relate image contents to meaningful features and a collection of features into a possible set of rules for understanding the underlying processes in a complex system. The intent is to build models for dynamic processes through observation of real data or to learn about the parameters of a model through experimental process. This can be achieved through intelligent summaries—high-level, multiview, and compact representations—that are based on meaningful features computed from data content. These features can then be used to answer queries that will quantify variability and trends.

## Accomplishments

We have developed a framework to represent spatio-temporal images in terms of vector fields and blobs. Vector fields can be subsequently represented in terms of singularities of the field. Blobs are homogeneous regions that correspond to distinct structural patches. Blobs can exist in two-dimensional or volumetric datasets, and they can represent organization of nuclei in a cultured colony, homogeneous regions in geophysical data, and tumor

locations in data from magnetic resonance imaging (MRI) or computed tomography (CT), etc. We have focused on developing an algorithm for detection and segmentation of blobs in two-dimensional images observed with a confocal microscope. This is significantly more difficult than detection of blobs from three-dimensional datasets since additional constraining information is missing. The problem is further complicated because of nonuniformities in intensity, presence of random and speckle noise, and the fact that distinct blobs may overlap each other. We have developed a computational framework that is layered and involves filtering for noise and internal substructures, followed by grouping of iso-intensity pixels into their local centroid. In this system, noise and internal substructures are detected with elliptic features. These elliptic features are then removed and interpolated with harmonic cuts. Harmonic cuts are equivalent to solving the Laplace equation on a multi-connected domain with irregular boundaries. Interpolation removes speckle noise and produces a smooth surface for the interior region of each blob. Objects that are touching one another are grouped according to their centroid and partitioned from the computed vector field.

The proposed technique is model based, assumes an ellipsoidal model for the gross shape of the blob (nuclei), which is not expressed as a parametric model, and does not rely on the user to provide an initial seed point for initial segmentation. Segmentation results for two touching nuclei and a collection of them in tissue are shown in Figures 16 and 17.

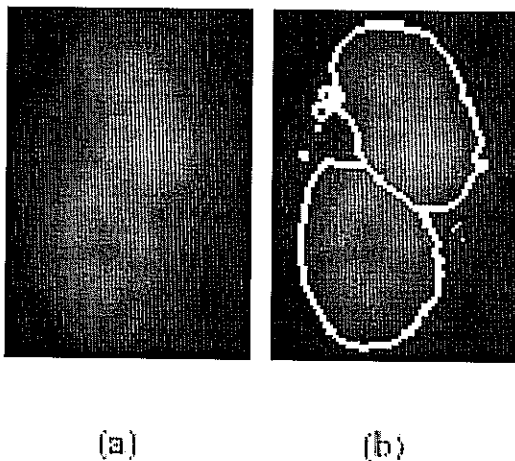


Figure 16: Segmentation of two touching nuclei.

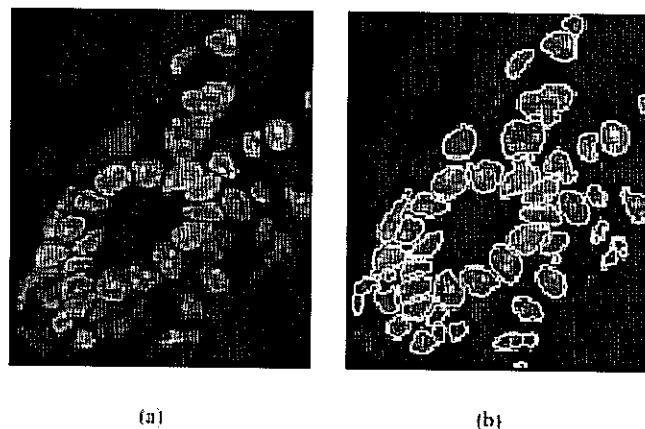


Figure 17: Detection and segmentation of blobs in tissue.

## Publications

Q. Yang and B. Parvin, "Harmonic Cut and Regularized Centroid Transform for Localization of Subcellular Structures," *Institute of Electrical and Electronics Engineers Conference on Computer Vision and Pattern Recognition – Sketch Series* (November 2001).

Q. Yang and B. Parvin, "Convex Hull of Elliptic Features for Blob Detection," submitted to *International Conference on Pattern Recognition*, Quebec City, Canada (August 2002).

Q. Yang and B. Parvin, "Harmonic Cut and Regularized Centroid Transform for Localization of Subcellular Structures," submitted to *Institute of Electrical and Electronics Engineers Transactions on Pattern Analysis and Machine Intelligence*.

Q. Yang and B. Parvin, "High Resolution Reconstruction of Sparse Spatio-Temporal Data," submitted to *Institute of Electrical and Electronics Engineers Transactions on Image Processing*.

---

## Linear Algebra and Statistical Algorithms for Text Classification in Large Databases

---

Principal Investigators: Horst Simon

Project No.: 99014

## Project Description

We continued our research in the areas of developing mathematical and statistical models/methods for information retrieval, text clustering/classification, and collaborative filtering. In order to accomplish these objectives, we also continued developing and implementing fast scalable parallel algorithms, especially algorithms for computing large-scale singular value decomposition used in the above-mentioned applications on high-performance computers.

The unique feature of this research is the integration of theoretical investigation based on statistics and matrix theory; algorithmic development using computational science methodologies and experimentation on real-world text corpora obtained from World Wide Web search engines. The research was carried out as a collaboration of

the National Energy Research Scientific Computing Center (NERSC) staff at Lawrence Berkeley National Laboratory with Professors Hongyuan Zha at Pennsylvania State University and Ming Gu at University of California, Berkeley. Through collaboration with Alexa (a subsidiary of Amazon) we have assured access to large text databases derived from their web crawls in 1998 and 1999. All production software development has been carried out on the NERSC Cray T3E and IBM SP. Both compute and input/output (I/O) capabilities of large parallel supercomputers are needed to effectively analyze document collections consisting of millions of pages.

## Accomplishments

There were several major accomplishments this year. In the area of web graph analysis we focused on the following three topics:

- The first is a better theoretical understanding of some of the issues impacting latent semantic indexing (LSI) performance on large, multi-topic document collections. We previously found that discriminating but rare terms are "lost" in the LSI representation of documents and queries. Using results from the perturbation theory of eigenvalue problems we can now explain why this occurs.
- Our second major accomplishment was an analysis of the "Hub and Authority" formulation used in World Wide Web searches. This analysis was validated with experiments on real data from the Web, ranging from small collections of pages to a site-level graph of the entire Web. We found that pages with high connectivity often dominate the rankings obtained by such methods.
- We have also been exploring the use of other techniques, both graph and linear algebra based, in an attempt to determine fundamental properties of the web graph. Preliminary results indicate that we can distinguish among different types of sites (such as home page repositories) and analysis is ongoing.

In the area of clustering algorithms the following major accomplishments were achieved:

- We described a new algorithm based on non-linear scaling for principal component analysis (PCA). The nonlinear scaling accelerates the self-aggregation of data into clusters. We tested this algorithm on gene expression profiles, web hyperlink structures, and internet newsgroup clustering. The new algorithm performed well, and provided sharp distinction between clusters.

- We proposed a new data clustering method based on partitioning a bipartite graph. This is applicable to many data types arising from data mining, e.g., term-document data sets. The partition is constructed by minimizing a normalized sum of edge weights between unmatched pairs of vertices of the bipartite

graph. An approximate solution of the minimization problem can be obtained by computing a partial singular value decomposition (SVD) of the associated edge-weight matrix of the bipartite graph.

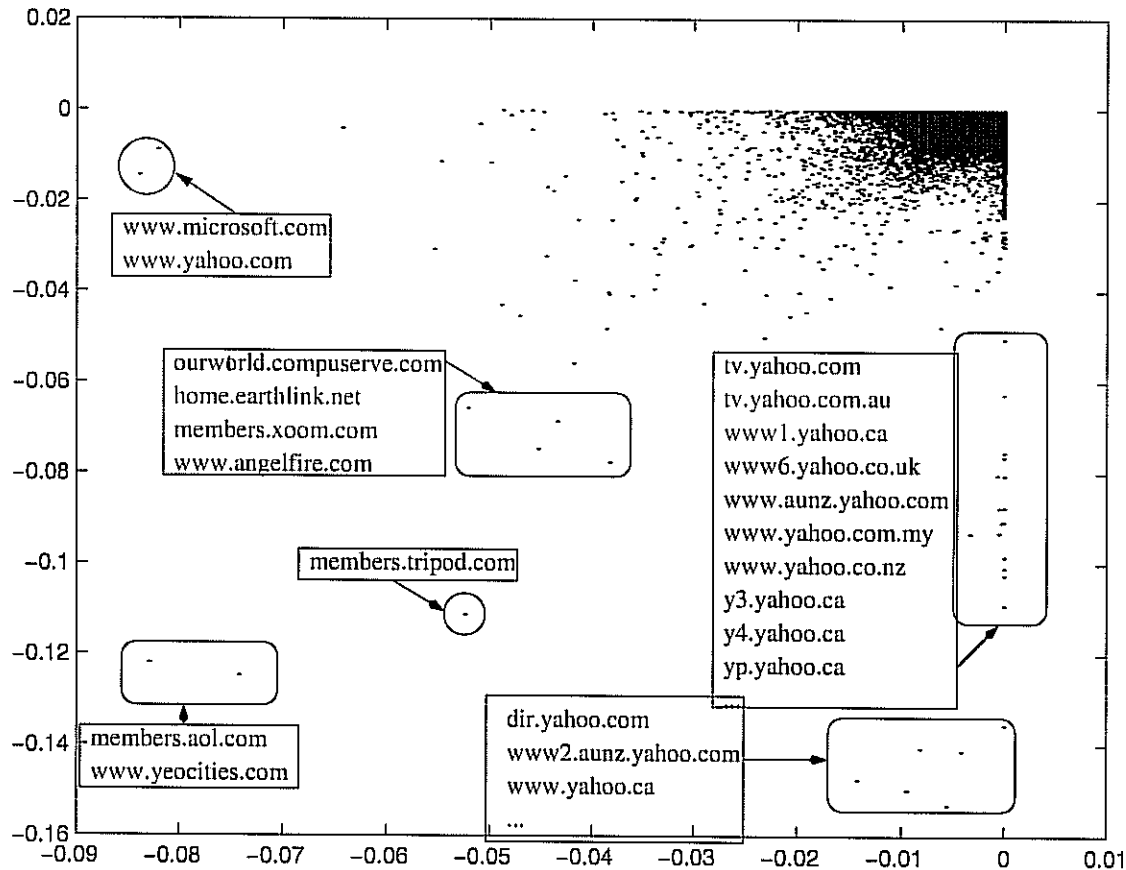


Figure 18: Hub weight vs. Authority weight for a site level graph of the World Wide Web. "Hubs" point to useful sources of information while "Authorities" are themselves informative sites. The calculation of these weights involves a large Singular Value Decomposition of the adjacency matrix of the web graph. The hub weights are identified with the first right singular vector ( $v_1$ ) and the authority weights with the first left singular vector ( $u_1$ ). It is interesting to note that some broadly related sites (directories, home page repositories, and other popular sites) can be grouped together using this technique.

## Publications

P. Husbands, H. Simon, and C. Ding, "On the Use of the Singular Value Decomposition for Text Retrieval," First Computational Information Retrieval Workshop (October 22, 2000).

C. Ding, H. Zha, X. He, P. Husbands, and H. Simon, "Analysis of Hubs and Authorities on the Web," LBNL Technical Report LBNL-47847, (May, 2001).

P. Husbands and H. Simon, "Spectral Views of the World Wide Web," (in preparation).

H. Zha, C. Ding, M. Gu, X. He, and H. Simon, "Spectral Relaxation for K-means Clustering," *Neural Information Processing Systems, NIPS\*2001*, Vancouver, British Columbia, Canada (December 3-8, 2001).

C. Ding, X. He, and H. Zha, "A Spectral Method to Separate Disconnected and Nearly-Disconnected Web Graph Components," *Proceedings of 7<sup>th</sup> International Conference on Knowledge Discovery and Data Mining, KDD2001*, 275-280 (August 2001).



## Earth Sciences Division

---

### ***Computer Tools and Methods for Demonstrating a Robotic Ocean Carbon Observing System***

---

Principal Investigators: James Bishop

Project No.: 01018

#### **Project Description**

Understanding the exchanges of carbon between the atmosphere and ocean and the fate of carbon delivered to the deep sea is fundamental to understanding the ocean carbon cycle and to ocean carbon sequestration science. We propose development of methods to rapidly process the daily data streams of environmental data from the first robotic ocean carbon observers. In addition, we propose at-sea acquisition of image data on large particle size distributions and abundances. This will lay the foundation for our efforts to develop a method for robotic observation of carbon flux in the ocean. These efforts are made in anticipation of implementation of a much larger ocean carbon-observing network that will be operational by the end of the decade, requiring forefront techniques for observation, analysis, and interpretation of the ocean carbon cycle.

The major focus of this effort was to develop methods to rapidly process the daily data streams of environmental data from the robotic carbon observers, which were deployed in the subarctic North Pacific in April 2001.

#### **Accomplishments**

##### ***Real Time Web-Based Processing of Data Streams from Robotic Sounding Oceanographic Lagrangian Observer (SOLO)-Carbon Explorer***

Our approach was to: (1) develop protocols for acquisition of data from the satellite operator on a daily basis; (2) cast the data into a geographic/temporal coordinate system, and physical/chemical units; (3) develop web protocols for the display of subsets of collected data; and (4) to begin to develop protocols for data interpretation and analysis by merging our observations with those of other remote sensing data such as surface solar irradiance and ocean biomass (including NASA's SeaWiFS and MODIS), and ocean circulation and winds (i.e. TOPEX/POSEIDON and QUIKSCAT).

Two SOLO-carbon Explorers have been operating in the North Pacific since April 2001. The geographic location and data from these observers is now available on the web in real time (<http://flameglo.lbl.gov/SOLO-robots.html>). Incoming data are automatically captured and transmitted to the web server for processing. Server software written in Java translates the data into physical units and renders information graphically in geographic and depth coordinate systems. Our system is designed to be easily scaled to allow real time display of information from two additional pairs of SOLOs to be deployed in the southern ocean in January 2002.

##### ***At-Sea Acquisition of Image Data of Sinking Marine Particles***

This acquisition of image data of sinking marine particles was done by using samples collected by the Multiple Unit Large Volume *in-situ* Filtration System (MULVFS) during a August 2001 cruise to the California current system. Laboratory Directed Research and Development (LDRD) supported the at-sea testing of a prototype imaging system constructed for autonomous assessment of large particles collected by an optical sediment trap. The at-sea test revealed a significant sensitivity to ship motion that had not been anticipated, and corrective measures have been applied.

---

### ***Carbon-Water-Climate Interactions***

---

Principal Investigators: Inez Fung, Horst Simon, Jim Bishop, and Don DePaolo

Project No.: 00009

#### **Project Description**

The goal of the project is to investigate the future co-evolution of carbon dioxide and climate using a coupled three-dimensional climate-biogeochemical model. Projections of future climate have relied on physical climate models where energy and water are exchanged among the atmosphere, land, ocean, and ice reservoirs. These models have not included interactive biogeochemistry where carbon uptake and release processes respond to the changing climate, and in turn alter the changing climate.

through their impact on and response to the abundance of carbon dioxide in the atmosphere.

Our Laboratory Directed Research and Development (LDRD) project starts with the implementation of interactive terrestrial and ocean carbon modules into the Community Climate System Model (CCSM), and implementation of the model on the National Energy Research Scientific Computing Center (NERSC)'s Cray Parallel Vector Processor (PVP) PVP cluster. An experiment will prescribe fossil fuel carbon dioxide emission rather than atmospheric carbon dioxide growth.

As the interaction of energy, water, and carbon are complex, a complex model such as ours is difficult to assess. To help constrain our interpretation of the feedbacks in the carbon-climate model, we have also initiated two new avenues of research to develop the auxiliary information and understanding to constrain the carbon-climate modeling. The first is the use of water isotopes to diagnose the sources and transformation processes for water. The second is in the area of aeolian deposition of iron and subsequent enhancement of marine productivity and drawdown of carbon dioxide from the atmosphere.

## Accomplishments

The land and ocean carbon modules have been successfully implemented in the National Center for Atmospheric Research (NCAR) CCSM-1. The terrestrial carbon module starts with stomatal conductance and gross photosynthesis calculated in the CCSM-1. Carbon is processed through the growth, death, and decay cycles of 14 different plant functional types within each model gridbox. The oceanic carbon module predicts carbon alkalinity, and the concentrations of dissolved inorganic and organic carbon, and phosphate in the ocean. Marine productivity (and hence particulate organic and inorganic carbon) is diagnosed as that required to bring the phosphate concentration to the observed level. A depth-dependent mineralization rate returns the particulate carbon to dissolved carbon species.

The control run for the fully coupled carbon-climate pre-industrial system is in the 50<sup>th</sup> year of integration. Thus far, the control remains stable, with little drift in the carbon system parameters and physical climate parameters. This is an important accomplishment, as natural background air-sea fluxes to and from the atmosphere are ~100 petagrams of carbon (PgC) per year. An imbalance of 1% per year over 250 years would be equal to the cumulative fossil fuel input since the pre-industrial era.

Our investigation into the water cycle has focused on the isotope exchange between raindrops and the ambient atmosphere. It is often assumed that isotopic equilibrium

between raindrops and the ambient atmosphere is fast, so that isotopic content of precipitation reflects that near the collector at the surface. Existing general circulation models, however, assume that precipitation from different cloud types have different "memories" of condensation conditions aloft, with the "memory degree" prescribed from ad-hoc assumptions. Our investigation starts with the observation that raindrop size increases with rain rate, and models the time for isotopic equilibration between a raindrop and the ambient atmosphere. The results quantify the equilibration time as a function of drop size and ambient temperature. With this investigation, we are deriving a totally new parameterization for the degree of isotopic equilibration based on rain rate. This parameterization will be implemented into the NCAR atmospheric general circulation model, which uses the distribution of water isotopes to diagnose the sources and transformations in the water cycle.

Our investigation into the dust cycle and its impact on atmospheric radiation, marine productivity, atmospheric carbon dioxide levels, and climate has been carried out with Drs. Pierre Biscaye and Aloys Bory of Lamont-Doherty Earth Observatory of Columbia University. Our previous modeling of dust and its impact on atmospheric radiation and on marine productivity required assumptions about dust composition, size distribution, and transport. To improve the models, we are obtaining new high-resolution data to constrain the modeling. The data are new dust samples with very high temporal resolution (to resolve seasonal variability over the last four years) from Greenland. The samples were melted and dust extracted by ultracentrifugation on site. Analysis of the new dust and meltwater samples for mineralogical and isotopic tracer characteristics is now in progress. Comparison of previously collected Greenland dust to dust-sized particles extracted from loess, desert sands, and arid soils from all over the northern hemisphere has shown eastern Asia to be the principal provenance region, although there is temporal variability in the source region. We have therefore initiated a modeling project to examine the seasonal and interannual variations of dust source regions and dust deposition. Unlike previous work, our work will include mineralogical as well as size information about the dust, and will capture the meteorological conditions of those years relevant for the uplift, transport, and deposition of the new dust samples. The new data and the model will yield new insights into how dust source regions, atmospheric abundance, and deposition may vary with climate. The information will be the first step towards an investigation into the dust-iron-carbon connection, where aeolian iron is thought to fertilize marine productivity and alter the abundance of atmospheric carbon dioxide.

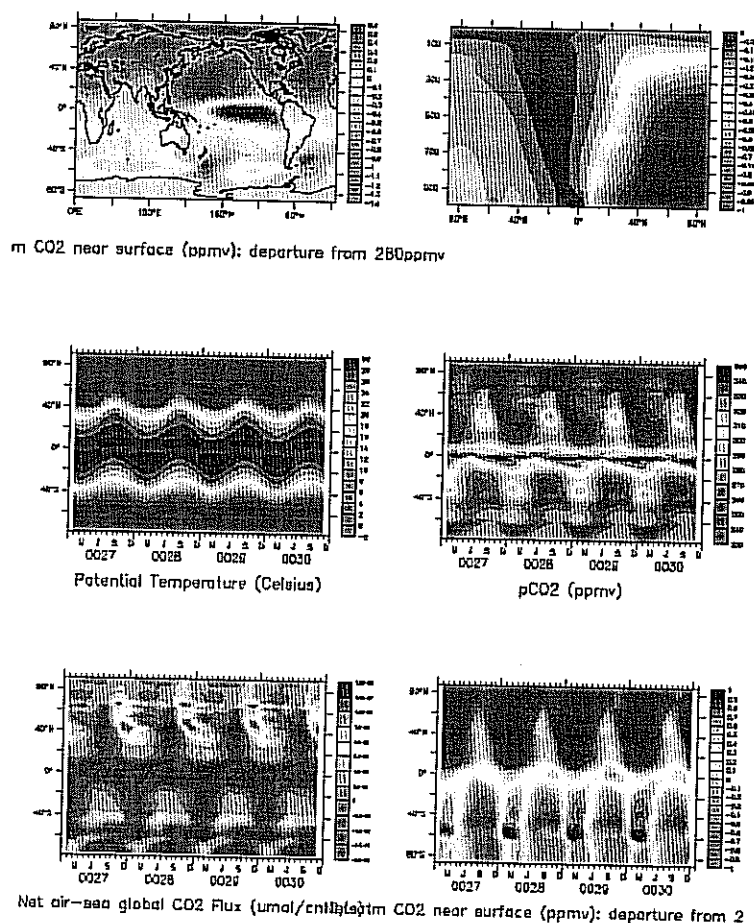


Figure 20: Results from years 25 to 30 of the coupled carbon-climate model. Upper panels: surface and meridional distribution of atmospheric carbon dioxide. Middle panels: Evolution of the zonally averaged distributions of surface temperature and near-surface pCO<sub>2</sub> in the ocean. Lower panels: Evolution of the zonally averaged distribution of air-sea carbon dioxide flux and near-surface carbon dioxide concentration in the atmosphere.

## Publications

M. Blackmon, *et al.*, "The Community Climate System Model," *Bulletin of the American Meteorological Society*, **82**, 2537-2376 (2001).

J.T. Randerson, C.J. Still, J.J. Ballé, I.Y. Fung, S.C. Doney, P.P. Tans, and J.W.C. White, "The <sup>13</sup>C Discrimination of Arctic and Boreal Biome Net CO<sub>2</sub> Exchange Inferred from Remote Atmospheric Measurements and Biosphere-Atmosphere Models," *Global Biogeochemical Cycles*, (2001) (in revision).

---

## ***Aerobic Bioremediation of Landfills***

---

Principal Investigators: Terry Hazen, Curtis Oldenburg, Sharon Borglin, and Peter Zawislanski

Project No.: 00010

### **Project Description**

The purpose of this research is to demonstrate and model aerobic bioremediation of municipal solid landfills as a more cost effective and environmentally sound way of managing municipal solid waste. This work is applicable not only to municipal solid waste, but also to remediation of solid waste landfills created by industry and the industrial processes of Department of Energy (DOE) and Department of Defense (DOD). The goals are to determine the critical biological and chemical parameters that control the ability of air injection and leachate recirculation in landfills to: (1) increase the biodegradation rate of the refuse mass, (2) decrease production of greenhouse gases ( $\text{CH}_4$ ), (3) regulate biogenic temperature, (4) reduce metals leaching, (5) increase short-term subsidence, and (6) increase the long-term stability of the refuse mass. An increased subsidence of only 15% could translate to over \$1 billion in additional revenues for the 3500 landfills currently in operation in the United States.

Using data collected over the last year from a demonstration conducted at the Columbia County, Georgia, Landfill, and a literature evaluation, critical biological, physical, and chemical parameters have been identified during the first year of the project. These potentially controlling parameters will be tested in the laboratory at Berkeley Lab using landfill simulation columns. The columns will be tested using various types of compaction, air injection strategies and rates, and leachate recirculation strategies and rates. During the first year, the landfill columns were designed and constructed and testing should begin shortly. The tests will be evaluated in relationship to the six goals stated above. These tests will be used to develop a numerical simulation model of aerobic bioremediation of landfills, building on the TOUGH2 code of Berkeley Lab. This approach will then be field demonstrated in the more arid environments at California municipal landfills and western DOE sites. Both the Yolo County and University of California at Davis landfills have been contacted.

### **Accomplishments**

Stabilization of municipal landfills is a critical issue involving land reuse, leachate treatment, and odor control. In an effort to increase landfill stabilization rates and decrease leachate treatment costs, municipal landfills can be operated as active aerobic or anaerobic bioreactors. Rates of settling and biodegradation were compared in three different treatments of municipal landfill materials in laboratory-scale bioreactors. 55-gallon clear acrylic tanks were fitted with pressure transducers, thermistors, neutron probe access tubes, leachate recirculation systems, gas vents, and air injection ports. The treatments applied to the tanks were (a) aerobic (air injection with leachate recirculation and venting from the top); (b) anaerobic (leachate recirculation with passive venting from the top), and (c) a control tank (passive venting from the top and no leachate recirculation).  $\text{O}_2$ ,  $\text{CO}_2$  and  $\text{CH}_4$  concentrations were measured at the gas vent, and leachate was collected at the bottom drain. The water saturation in the aerobic and anaerobic tanks averaged 17% and the control tank averaged 1%. All tanks contained 10 centimeters of pea gravel at the bottom, overlain by a mixture of fresh waste materials on the order of 5 to 10 centimeters in size. Relative degradation rates between the tanks were monitored by  $\text{CO}_2$  and  $\text{CH}_4$  production rates and  $\text{O}_2$  respiration rates. Respiration tests on the aerobic tank show a decrease in oxygen consumption rates from 1.3 mol/day at 20 days to 0.1 mol/day at 300 days, indicating usable organics are being degraded. The anaerobic tank produced measurable methane after 300 days that increased to 41% by volume after 370 days. Over the test period, the aerobic tank has shown 30% settling, the anaerobic tank 18.5% settling, and the control tank 11.1% settling. The leachate in the aerobic tank has shown concentrations of metals, nitrate, phosphate, and Total Organic Carbon (TOC) an order of magnitude lower than the leachate from the anaerobic tank.

In FY 2001, we developed and tested a module (T2LBM) for the TOUGH2 simulator that implements a Landfill Bioreactor Model to provide simulation capability for the processes of aerobic or anaerobic biodegradation of municipal solid waste and the associated flow and transport of gas and liquid. While prior simulation models were either batch models (zero-dimensional), multilayer batch models (one-dimensional), or did not consider gas production as part of biodegradation, T2LBM is fully three-dimensional and models gas and liquid flow in addition to biodegradation of acetic acid by either aerobic or anaerobic microbial activity. The enormous complexity of the landfill environment necessitated some simplifications.

The approach chosen for T2LBM was to couple the process modeling of the flow and transport of gas and aqueous phases inherent in TOUGH2 with new biodegradation and

gas generation process capability. For simplicity, T2LBM models the biodegradation of a single substrate component (acetic acid,  $\text{CH}_3\text{COOH}$ ) as a proxy for all of the biodegradable fractions in municipal solid waste. T2LBM includes six chemical components ( $\text{H}_2\text{O}$ ,  $\text{CH}_3\text{COOH}$ ,  $\text{CO}_2$ ,  $\text{CH}_4$ ,  $\text{O}_2$ ,  $\text{N}_2$ ) and heat distributed in gaseous and aqueous phases with partitioning by Henry's law. This approach assumes implicitly that hydrolysis reactions occur to produce acetic acid, and places the model focus on biodegradation and associated gas production along with flow and transport processes including non-isothermal

effects. We have tested T2LBM against results from published laboratory studies of aerobic and anaerobic biodegradation. In addition, we have simulated a hypothetical two-dimensional municipal solid waste landfill with leachate recirculation and subsequent air injection into the bottom of the landfill. These demonstrate code capabilities including gas production, and evolution of acetic acid and oxygen concentrations, temperature, aerobicity, and pH. We have also begun computer simulations of the landfill bioreactor experiments being carried out in the lab component of this project.

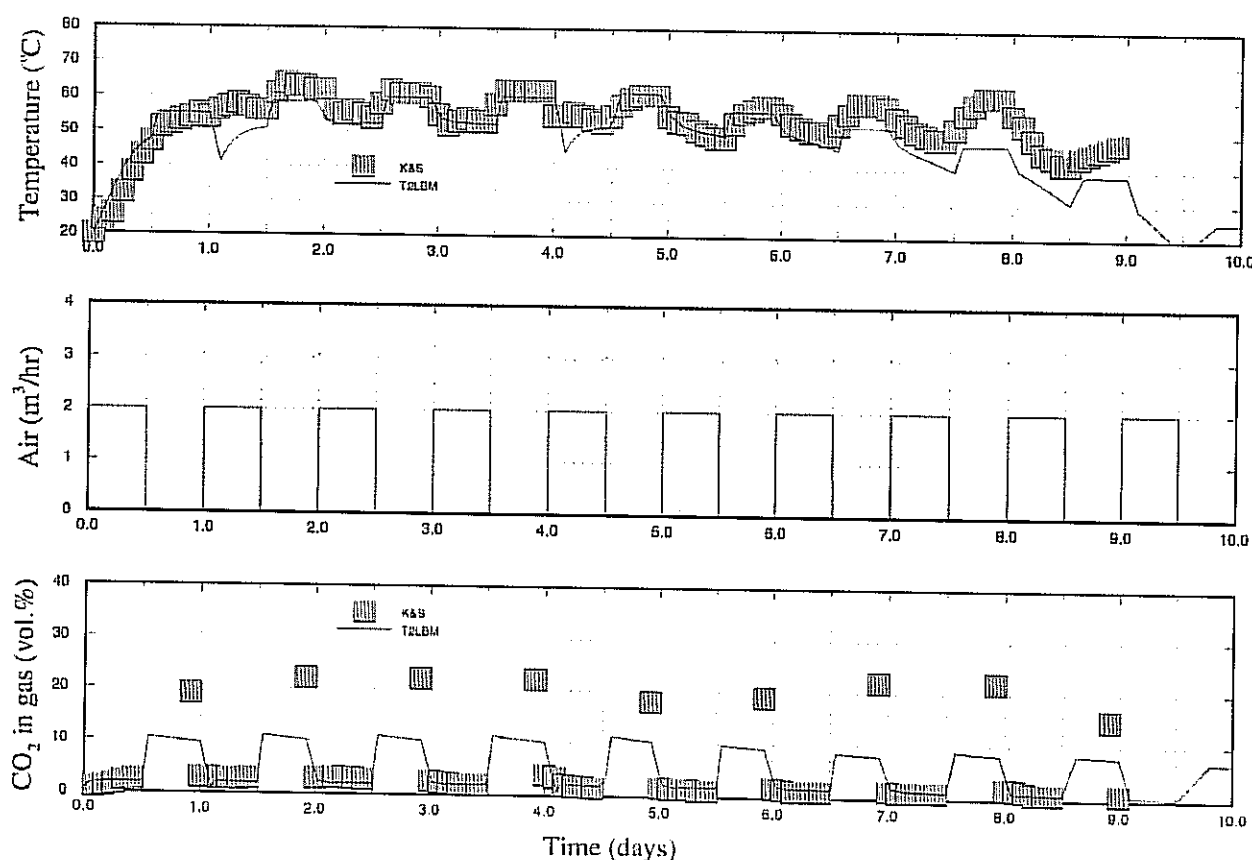


Figure 21: Landfill model T2LBM predictions vs. actual data comparisons.

## Publications

C.M. Oldenburg, "T2LBM: Landfill Bioreactor Model for TOUGH2, Version 1.0," Lawrence Berkeley National Laboratory Report LBNL-47961 (April 2001).

C.M. Oldenburg, S.E. Borglin, and T.C. Hazen, "Process Modeling of Flow, Transport, and Biodegradation in Landfill Bioreactors," *EOS, Transactions American*

*Geophysical Union, Fall Meeting* and Lawrence Berkeley National Laboratory Report LBNL-48926 (October 2001).

S.E. Borglin, T.C. Hazen, C.M. Oldenburg, and P.T. Zawislanski, "Mesoscale Laboratory Models of the Aerobic Biodegradation of Municipal Landfill Materials," *EOS, Transactions, American Geophysical Union, Fall Meeting* and Lawrence Berkeley National Laboratory Report LBNL-48926 (October 2001).

T.C. Hazen, "In-Situ Respiration and Direct Enzymatic Assays for Assessing Bioremediation," *EOS, Transactions, American Geophysical Union, Fall Meeting* and Lawrence Berkeley National Laboratory Report LBNL-48927 (October 2001).

---

## **Geological Sequestration of Carbon Dioxide**

---

Principal Investigators: Larry Myer and Sally Benson

Project No.: 00008

### **Project Description**

The climate of the earth is affected by changes in radiative forcing due to several sources including greenhouse gases ( $\text{CO}_2$ ,  $\text{CH}_4$ , and  $\text{N}_2\text{O}$ ). Energy production and the burning of fossil fuels are substantially increasing the atmospheric concentrations of  $\text{CO}_2$ . One of several proposed strategies to reduce atmospheric emissions is to capture  $\text{CO}_2$  from fossil fuel power plants and sequester it in the deep underground.

The Department of Energy (DOE) is addressing sequestration through a broad program with the goal of developing "safe, predictable and affordable ways to prevent  $\text{CO}_2$  from building up in the atmosphere" by 2025. The goals of this Laboratory Directed Research and Development (LDRD) were to pursue research on key scientific issues concerning mechanisms for subsurface trapping of  $\text{CO}_2$  and monitoring of  $\text{CO}_2$  migration with the goal of establishing Berkeley Lab's scientific leadership in this area.

Two projects were carried out: (1) evaluation of the sensitivity of seismic and electromagnetic geophysical techniques for detecting the pressure of  $\text{CO}_2$  in the subsurface, and (2) development of a new well pressure testing technique for monitoring the progress of  $\text{CO}_2$  injection into the subsurface.

The technical approach involved analytic and numerical simulation, and laboratory measurements. The sensitivity of geophysical techniques was evaluated through laboratory measurements of seismic and electrical properties on rock cores containing brine and  $\text{CO}_2$ . Numerical simulations involved using the TOUGH2 code to investigate pressure build-up and fall-off in heterogeneous reservoirs, while a new approximate analytical solution was developed for calculating pressure build-up at  $\text{CO}_2$  injection wells.

### **Accomplishments**

#### *Laboratory Measurements of Seismic and Electric Properties*

Geophysical techniques provide the most cost-effective approach for obtaining the spatial coverage required for mapping the location and movement of  $\text{CO}_2$  in the subsurface. However, the effectiveness of these techniques depends upon many factors. Contrasts in physical properties such as seismic wave velocity and electrical conductivity are of first-order importance, though only limited laboratory data are available to define these properties.

The objective of the laboratory measurements is to obtain concurrent seismic and electrical properties of rock under reservoir conditions containing  $\text{CO}_2$ , brine and oil. The first task was to assemble apparatus, which required modification of a test cell and fabrication of transducers. Samples measuring 1-1/2 inches in diameter by 3 inches long are placed in a rubber jacket, which contains four equally spaced electrodes in contact with the rock. Seismic transducers are mounted at the ends of the sample for pulse transmission tests. Each transducer contains piezoelectric elements with a center frequency of 500 kHz.

Test conditions were applied so that electrical and seismic properties could be measured as the  $\text{CO}_2$  passed from the gas phase through the critical point to liquid-like properties. Effective stress conditions were representative of moderate depth reservoir conditions. Measurements were made at a confining pressure of 1500 pounds per square inch (psi). Measurements were first made under brine-saturated conditions, beginning with a pore pressure of 300 psi and increasing to about 950 psi. The pore pressure was then reduced to 300 psi and  $\text{CO}_2$  was injected. The  $\text{CO}_2$  was flushed through the core against a backpressure of 300 psi until residual brine saturation was achieved. At this point the outlet valve was closed and the brine saturation held constant while the  $\text{CO}_2$  pressure was increased. Tests were conducted on a sample of Berea sandstone and on a sample of diatomite from the Lost Hills, California, oil reservoir. Results from the sandstone are presented first.

Berea has a porosity of about 20% and a permeability in the range of 150 to 300 millidarcies. The electrical resistivity of the sandstone saturated with 1.1 ohm-meter ( $\Omega\text{-m}$ ) brine was about 17  $\Omega\text{-m}$  and when saturated with 3.5  $\Omega\text{-m}$  brine about 43  $\Omega\text{-m}$ . The resistivity was constant for each brine concentration over the range of pore fluid pressures used in the tests.

Introduction of  $\text{CO}_2$  caused a significant increase in resistivity of the sample. The resistivity of the sample in the test using 1.1  $\Omega\text{-m}$  brine increased from 17  $\Omega\text{-m}$  to 38  $\Omega\text{-m}$ , while that for the test using 3.5  $\Omega\text{-m}$  brine increased from 43  $\Omega\text{-m}$  to about 80  $\Omega\text{-m}$ . Increasing the pressure of the

CO<sub>2</sub> in the gaseous phase caused an increase in sample resistivity. Behavior above the gas/liquid phase line differed for the different brine concentrations. For the 1.1 Ω-m brine test the resistivity at 900-psi pore pressure (in the liquid phase) was about equal to that at 800 psi. The 3.5 Ω-m brine test showed a decrease in resistivity between 800-psi pore pressure and 950 psi. Further testing needs to be done to evaluate changes in resistivity in the region of the critical point of CO<sub>2</sub>.

When fully saturated by brine, increasing the pore pressure from 300 psi to 950 psi caused a 6% reduction in P-wave velocity. Injection of CO<sub>2</sub> caused a significant reduction in P-wave velocity. At 300 psi pre-pressure, flushing the sample with gaseous CO<sub>2</sub> caused an 11% reduction in velocity. This reduction was due to replacement of the liquid brine by low density, low modulus gas phase CO<sub>2</sub>. The further reduction caused by increasing CO<sub>2</sub> pressure to 600 psi was similar to that observed for the brine saturated test and is therefore considered to be due to a reduction in effective stress.

The reduction upon pressurization to 800 psi was more than would be expected from effective pressure changes. It is consistent with a rapid reduction in the velocity of CO<sub>2</sub>, which has been observed near the critical point. At 900-psi pore pressure the velocities recover, reflecting the change to liquid-like properties. The velocity is less than that for brine-saturated conditions because of the lower density and bulk modulus of the CO<sub>2</sub>.

Measurements on the diatomite sample were made to support field tests of crosswell and single well seismic, and crosswell electromagnetic (EM) methods, which are being developed and evaluated as monitoring techniques. An early opportunity to begin field evaluation of these technologies has been provided by a pilot CO<sub>2</sub> flood being conducted by Chevron in the Lost Hills oil field. The pilot targets the producing interval in the Belridge Diatomite, a member of the Monterey Group. The top of the diatomite is at a depth of about 1400 feet and extends to a depth of about 2100 feet. The diatomite reservoir rock is characterized by high porosity (40% to 60%) but low permeability (<1 to 10 millidarcies). Measurements were made on a sample of diatomite from the reservoir interval. Test procedures were the same as those used for the sandstone sample.

The diatomite sample showed similar trends as the sandstone, that is, as CO<sub>2</sub> was injected into the rock, seismic velocity decreased and resistivity increased. Interpretation of field measurements often requires use of empirical correlations between different types of well logs. One correlation, which is frequently used, is sonic velocity versus electrical resistivity. The laboratory measurements showed a linear trend of decreasing P-wave velocity with increasing electrical resistivity.

Results of the laboratory measurements showed that both seismic and electrical geophysical methods should be useful for detecting CO<sub>2</sub> in the subsurface. Further work is needed to define quantitative rock physics relationships, particularly in the region of the critical point of CO<sub>2</sub> and when mixed phases are present.

#### *Well Pressure Testing for Monitoring of Carbon Dioxide*

Observation of pressure build-up and fall-off in injection wells is one of the common methods for monitoring the progress of an injection operation. Evaluation of these data can be used to detect changes in the near-bore permeability and in some cases, to track migration of the flood front. Reservoir heterogeneity and gravity override may influence the applicability of the methods used for interpreting these parameters, particularly in the case of CO<sub>2</sub>, which is considerably less viscous and less dense than the resident formation fluids. This study used the numerical simulator TOUGH2 to investigate pressure build-up and fall-off responses in heterogeneous brine-filled reservoirs. These results were compared to pressure build-up and fall-off for calculating pressure buildup at CO<sub>2</sub> injection wells. Based on these comparisons, conclusions were made regarding the effect of formation heterogeneity and gravity override on the pressure transient responses and the extent to which these data could be valuable for monitoring the progress of CO<sub>2</sub> injection operations.

#### **Publications**

L.R. Myer, "Laboratory Measurements of Geophysical Properties for Monitoring of CO<sub>2</sub> Sequestration," *Proceedings of First National Conference on Carbon Sequestration*, Washington, D.C. (May 14-17, 2001).

S.M. Benson, "Effects of Reservoir Heterogeneity on Pressure Build-Up and Fall-Off at CO<sub>2</sub> Injection Wells," American Association of Petroleum Geologists Annual Convention, Denver, Colorado (June 3-6, 2001).

---

## ***Isotopic Analysis of Carbon Cycling and Sequestration in a California Grassland under Global Change Conditions***

---

Principal Investigators: Margaret Torn

Project No.: 00011

### **Project Description**

This is a project to develop advanced isotopic techniques to quantify carbon fluxes and storage in terrestrial ecosystems, and apply them to a California global change experiment. Field experiments combined with measurements of  $^{13}\text{C}$  and  $^{14}\text{C}$  will be used to study soil respiration, decomposition, and long-term carbon stabilization. The primary research questions are (1) How do changes in climate and nutrients influence carbon loss through respiration by plants and microbes? and (2) How does soil mineralogy influence the response of soils to global change and their capacity to stabilize organic carbon for long-term storage? The research will be expanded to address the question (3) How do changes in climate and nutrients influence microbial communities and the pools of carbon available for decomposition by different microbial communities?

The approach employs specialized field sampling and rapid, automated isotope analysis being developed under this Laboratory Directed Research and Development (LDRD) project. The Jasper Ridge Global Change field experiment, in an annual grassland, is the first to address the integrated suite of environmental changes predicted to occur over the coming decades: elevated  $\text{CO}_2$ , warming, increased rainfall, and nitrogen fertilization. The elevated  $\text{CO}_2$  creates isotopically labelled plant material that is used to quantify carbon fluxes and sources. The isotopic content of soil respiration, soil gas, and dissolved inorganic carbon are used to estimate the relative contribution to effluxes from recent plant photosynthate vs. decomposition of older soil organic matter. The influence of soil mineralogy on sandstone and serpentine soils is being tested by analyzing physically (density) fractionated soil for carbon and carbon isotopic content. This approach amplifies the signal of changes in carbon stocks and of mineral associations.

We have also applied the specialized  $^{13}\text{C}$  and  $^{14}\text{C}$  methods to soils from a Russian soil archive.

### **Accomplishments**

#### ***Effect of Global Change Treatments on Soil Carbon Losses from Microbial and Plant Respiration***

Global atmospheric change may have different impacts on microbial decomposition of organic matter (OM) compared to root respiration and exudates, but quantifying the cycling of different C sources through soil is difficult. We used the  $^{13}\text{C}$  isotopic tracer provided by elevated  $\text{CO}_2$  experiments to explore the effects of multiple global change treatments on soil respiration derived from roots, recent inputs, and from the older OM that represents long-term carbon sequestration in soil. We found that decomposition of older organic matter was accelerated by N fertilization but was not affected by the climate change (warming, water) treatments. Nitrogen fertilization also increased plant productivity, which could in principal compensate for the increased soil C losses. However, our research also showed that the recent plant inputs in the nitrogen treatments were rapidly respired from the soil rather than becoming stabilized in microbial biomass or soil organic matter.

#### ***Effect of Elevated $\text{CO}_2$ on Soil Carbon Storage and Turnover***

A number of recent studies have suggested that grassland soils may act as modest sinks for atmospheric  $\text{CO}_2$  as its concentration rises. However, detection of soil C sequestration in  $\text{CO}_2$  experiments is hampered by large, spatially variable C pools that turn over slowly compared to the duration of experiments. We used stable isotopes and density separation techniques to isolate and investigate changes in the size of intermediate-cycling (light fraction) and slow-cycling (heavy fraction) soil C pools in two types of California grassland ecosystems, sandstone and serpentine, exposed to six years of elevated  $\text{CO}_2$ . The  $\delta^{13}\text{C}$  results showed that while only 3% to 8% of heavy fraction (HF) C turned over during the course of the experiment in the two grasslands, 21% to 36% of the light fraction (LF) C turned over, providing a strong signal to detect changes in pool size. Nevertheless, we detected no changes in soil C in either soil fraction of either grassland, indicating that the modest increases in annual ecosystem C uptake observed in this study did not lead to additional C storage.

To improve resolution of soil organic matter (SOM) dynamics, we used the dual  $^{13}\text{C}$  and  $^{14}\text{C}$  signatures of the fossil experimental  $\text{CO}_2$  to model turnover times of distinct C pools within each fraction. In both sandstone and serpentine soils, we found that only 20% to 40% of the LF had turnover times <10 years, while the rest of the LF had turnover times >100 years. We showed that the fast-cycling portion of the LF turned over completely during the 6-year experiment, accounting for the 21% to 36% observed turnover of C in the LF. The <1% of total soil C that makes up this fast-cycling pool sets an upper limit on the amount



of total soil C that can be lost or gained rapidly. Our findings illustrate that the potential for changes in soil C storage on time scales of years to decades, in these grasslands, is extremely limited, regardless of increases in ecosystem C uptake.

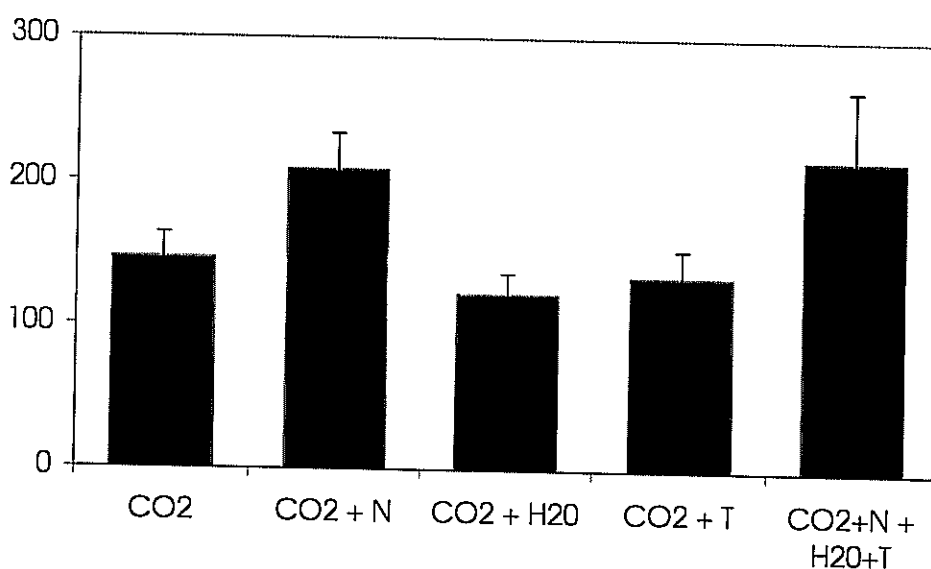
#### *Applying Dual Isotope Signatures to Historic Russian Soils*

We characterized soil carbon dynamics in a Russian steppe preserve using a 100-year-old soil archive and modern samples collected from the same site. In the preserve, the amount of organic carbon did not change appreciably between the 1900 and 1997 sampling dates, with 32 kilograms C/m<sup>2</sup> in the top meter and a third of that in the top 20 centimeters. Carbon and nitrogen stocks varied by less than 6% between two replicate modern soil pits or between the modern sites and the archive. Radiocarbon content decreased with depth in all sites and the modern SOM had positive  $\Delta^{14}\text{C}$  values near the surface due to nuclear weapons testing in the early 1960s. In the upper 10 centimeters, most of the SOM had a turnover time of 6 to 10 years, according to a model fit to the radiocarbon content. Below about 10 centimeters, the organic matter was almost all passive material with long (millennial) turnover times. Soil respiration  $\Delta^{14}\text{CO}_2$  on a summer day was 106‰ to 109‰, an isotopic disequilibrium of about

9‰ relative to atmospheric  $^{14}\text{CO}_2$ . In both the modern and archive soil, the relative abundance of  $^{13}\text{C}$  in organic matter increased with depth by 2‰ in the upper meter from  $\delta^{13}\text{C}$  equals -26 ‰ at 5 centimeters to -24‰ below a meter. In addition, the slope of  $\delta^{13}\text{C}$  vs. depth below 5 centimeters was the same for both soils. Given the age of the soil archive, these results give clear evidence that the depth gradients are not due to depletion of atmospheric  $^{13}\text{CO}_2$  by fossil fuel emissions but must instead be caused by isotopic fractionation between plant litter inputs and preservation of SOM. Overall, the data show that these soils have a large reservoir of recalcitrant C and stocks had not changed between sampling dates 100 years apart.

#### *Effect of Global Change Treatments on Microbial Community: Phospholipid Fatty Acids*

For the microbial community analysis (Phospholipid Fatty Acids analysis, PLFA): we developed the laboratory capability to handle the large numbers of samples, ran test samples, and made modifications to the extraction procedure for the Global Change experiment's grassland soils. We collected and preserved samples from a full factorial of global change treatments (128 samples for each of two years). We will complete the PLFA analysis of samples by June 2002.



*Figure 22: Annual soil respiration from microbial decomposition of soil organic matter under global change treatments. Nitrogen fertilization (CO<sub>2</sub> + N or CO<sub>2</sub> + N + watering + warming) was linked to increased losses of carbon from soil. Isotopic analysis of  $^{13}\text{CO}_2$  was used to quantify the fraction of respiration coming from decomposition of older organic matter, versus that from plant respiration and exudates.*

## Publications

M.S. Torn and J. Southon, "A New  $^{13}\text{C}$  Correction for Radiocarbon Samples from Elevated- $\text{CO}_2$  Experiments," accepted for publication in *Radiocarbon*.

M.S. Torn, A.G. Lapenis, A. Timofeev, M. Fischer, I. Babikov, and J. Harden, "Organic Carbon and Carbon Isotopes in Modern and 100-Year-Old Soil Archives of the Russian Steppe," accepted for publication in *Global Change Biology*.

E.S. Zavaleta, M.S. Torn, and C.B. Field, "Soil Carbon Storage and Turnover Responses to Elevated  $\text{CO}_2$  in Two Annual Grassland Ecosystems," in preparation for *Geophysical Research Letters*.

S. Davis, M.S. Torn, A. Phillips, and F. Fourmel, "Improving the Accuracy of Stable Isotope Analysis of Atmospheric and Soil Trace Gases Using Automated Multiple Sampling Combined with Standard Trace Gas Pre-concentration System," 222<sup>nd</sup> American Chemical Society National Meeting, Chicago, IL (August 26-30, 2001).

M.S. Torn, M.R. Shaw, C.B. Field, and H. Mooney, "Variation in Decomposition and Root Respiration Seasonally and Under Simulated Global Change in a California Grassland," American Geophysical Union Fall Meeting EOS Supplement (December 2000).

# Engineering Division

---

## ***Fast Lipoprotein Identification Process***

---

Principal Investigators: Henry Benner and Ronald Krauss

Project No.: 01040

### **Project Description**

The measurement of lipoprotein (LP) particles in human blood is used to assess the risk of coronary heart disease (CHD). Risk is related to the size distribution and cholesterol content of lipoprotein particles. With appropriate analytical methodology, LP is classified by particle size into several well-known categories such as high density lipoprotein (HDL) and low density lipoprotein (LDL) along with additional less well-known classes and subclasses. This project evaluates an aerodynamic mobility technique for measuring the size distributions of LP for the purpose of designing a faster, more accurate, and cheaper analytical method for LP. Gel electrophoresis is the most commonly used technique for revealing LP classes but it is slow and requires a well trained operator and data interpreter. We have determined that aerodynamic electrical mobility measurements can be used to display LP size distributions in about five minutes compared to a day or two for gel separations. Detecting mobility classified LP is based on nucleated condensation and allows LP concentrations to be determined accurately, thus circumventing the chromogenic problems encountered when lipid or protein stains are used to measure LP in gels. We have demonstrated that high-risk patients can be distinguished from low-risk patients on the basis of LP spectra measured with aerodynamic mobility.

### **Accomplishments**

Plasma was obtained from a number of patients enrolled in a cholesterol study. The LP profile of the patients was first evaluated with classical techniques and used as a criterion to select a small subset of patients whose LP profiles provided a wide range of patterns for investigation with the mobility technique. Total LP was separated from plasma using ultracentrifugation. The resulting LP was desalted and diluted and analyzed using a combination of electrospray ionization, differential mobility separation, and nucleated condensation to generate size spectra of LP particles in the 5 to 100 nanometers diameter range.

Aerodynamic electrical mobility analysis is based on measuring the drift of charged particles as they are dragged through air by the force of an electric field. The measurement process begins by aerosolizing LP into a stream of air using the electrospray process. Electrospray initially generates highly charged particles and in order to simplify the spectra the particles were converted to singly charged species. The singly charged particles were subjected to mobility analysis by passing them through a differential mobility analyzer, which provides a means for converting drift velocity into particle diameter. For this application, particle size spectra were obtained by scanning across the range of electrical mobility that corresponds to the 5 to 100 nanometers range. Peaks in particle size spectra revealed the concentration of particles in specific LP classes and the modal size of each distinguishable class. We were able to distinguish high-risk patients from low-risk patients on the basis of LP size distribution spectra.

Additionally, we showed that LP concentration measurements were linear over three orders of magnitude. When compared to concentration measurements of LP in gels, LP staining is known to vary with particle size because the lipid/protein content of LP changes with size. Our nucleated condensation detection scheme for LP particles forces vapor to condense onto individual particles and grow into "fog" droplets that are efficiently detected by laser light scatter. Our size distribution spectra can be plotted on a number basis (number of particles in a size bin vs. particle diameter) or converted to a mass basis and plotted as mass of LP per size bin vs. particle diameter. The mass vs. diameter spectra define the physical size distribution of the LP particles and are free from artifacts introduced specifically by LP staining techniques.

Finally, the size distributions obtained with mobility measurements show the presence of LP particles larger than LDL (5 to 30 nanometers range). Such particles are known as very low density lipoprotein (VLDL) and are not easily measured with gel electrophoresis. VLDL particles are thought to provide additional support for predicting CHD since there is recent and growing evidence for the role of VLDL in CHD. It appears that aerodynamic electrical mobility measurements will also provide a routine method for obtaining VLDL quantitation and size distributions.

### **Publications**

W.H. Benner, R.M. Krauss, and P.J. Blanche, "Ion Mobility of Biological Particles," patent applied for, 2001.



# Environmental Energy Technologies Division

---

## **Development of Cool Colored Shingles**

---

Principal Investigators: Hashem Akbari and Paul Berdahl

Project No.: 01019

### **Project Description**

This project is designed to stimulate the development of more reflective roofing materials that would mitigate urban heat island temperature buildup and thus reduce the formation of urban smog and the use of air conditioning. Cool roofing products are generally not available for the residential sector. The solar reflectance of asphalt roofing shingles, the predominant roofing material used for residential buildings in the United States, is largely determined by the pigments in the coatings on the roofing granules used to surface the shingles. Since half of the heat from the sun arrives as invisible radiation in the near-infrared spectrum, it is theoretically possible to create a "cool" shingle of any color by using a color pigment with high infrared reflectivity.

We are investigating how the optical and other physical (size, shape, etc.) properties of pigment particles affect the solar reflectance of roofing shingles. In particular, we are measuring the spectral reflectances and transmittances of coatings with individual pure pigments and thereby deriving the Kubelka-Munk (KM) coefficients, which describe the absorption and scattering strengths of the pigments at each wavelength in the solar spectrum. The spectral reflectance of a coating made with a mixture of pigments can be computed from these KM coefficients.

### **Accomplishments**

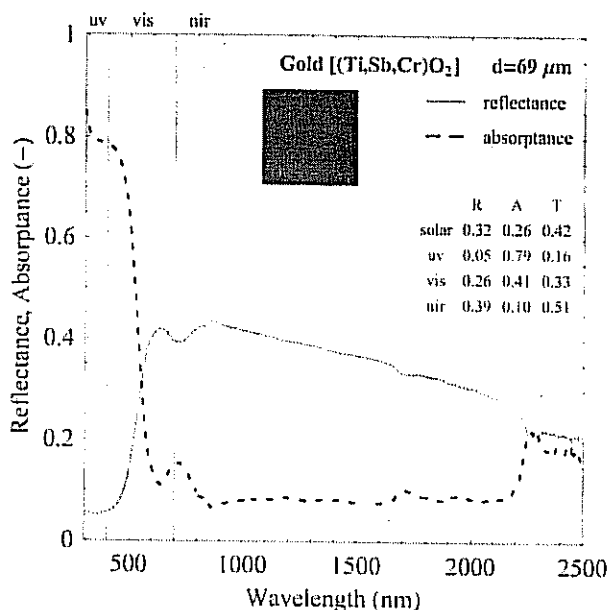
We have measured the spectral optical properties of many individual pigments, including conventional (e.g., chromium-oxide green) and "cool" (e.g., infrared-reflective black) pigments, by preparing coatings with single pigments (masstones) and mixtures with white (tints) on transparent glass slides. Diffuse transmittance and reflectance were measured as a function of wavelength across the solar spectrum. Diffuse absorptance was computed by subtracting the sum of the transmittance and reflectance from unity. Then, a detailed mathematical analysis of the data yielded Kubelka-Munk coefficients

that describe the optical scattering ( $S$ ) and absorption ( $K$ ) coefficients of the pigments. The measurements thus characterized the spectral radiative properties of various specific pigments (see Figure).

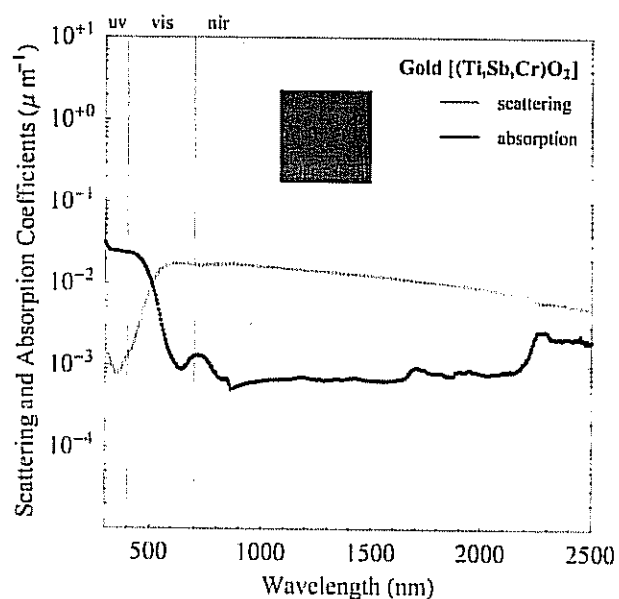
In general, the suitability of pigments for formulation of cool colored coatings is determined by the values of the absorption coefficient  $K$  in the near infrared. For example among black pigments, we learned that carbon black,  $\text{Fe}_3\text{O}_4$ , and  $\text{CuCr}_2\text{O}_4$  are not suitable. Nevertheless, certain mixed-metal oxides are suitable blacks. These materials include  $(\text{Cr}, \text{Fe})_2\text{O}_3$  (Cr:Fe ratio, 100:1) doped with other metal oxides, and proprietary mixed metal oxides. Black pigments are often used in color recipes, so it is critical to avoid the use of warm black pigments.

We have developed a computer program that estimates the spectral solar reflectance of coatings from the measured spectral absorption ( $K$ ) and scattering ( $S$ ) properties of pigments, and used this to compare the measured and predicted reflectances of mixtures and layers of pigments.

We have also filed a patent disclosure on a novel granule-coating technology. We have demonstrated the ability of this technique to increase the solar reflectivity of a "cool" blue coating while matching the original shade of blue.



(a)



(b)

Figure 19: Measured and computed properties of a gold-colored coating for shingle granules. Shown are (a) the spectral reflectance and absorptance of a 69 micron thick coating on transparent glass, and (b) the pigment's scattering and absorption coefficients computed from spectral reflectance and transmittance measurements.

## Publications

R. Levinson, P. Berdahl, and H. Akbari, "Solar-Spectrum Optical Properties of Pigments for Cool Roofing Materials," in preparation for LBNL Report.

H. Akbari and P. Berdahl, "Inexpensive Method for Producing Colored Coatings which Remain Cool in the Sun," Invention disclosure IB 1722, (September 2001).

## Atmospheric Chemistry: Changes at the Interface between Urban and Regional Scales

Principal Investigators: Ronald Cohen, Nancy Brown, Allen Goldstein, Robert Harley, and Haider Taha

Project No.: 00012

## Project Description

A critical challenge for the atmospheric sciences is to understand the anthropogenic impacts on atmospheric chemistry over spatial scales ranging from the urban to the regional, and, ultimately, to the global. Using Laboratory Directed Research and Development (LDRD) funding as a seed, we plan to conduct research focused on exploring, characterizing, and understanding these impacts. This problem is energy relevant because the majority of emissions that affect atmospheric change are combustion

generated. The problem is challenging due to the inherent non-linearity of the photochemistry at all spatial scales; the difficulties in simultaneously representing atmospheric fluid dynamics at different scales within a numerical model; the large demands placed on computational resources by solving the chemical rate equations; and the limitations of observations that are too sparse, infrequent, and inaccurate.

To address these issues, we propose to continue developing a combined experimental and modeling program whose objective is to understand: (1) the dependence of  $O_3$  sensitivity to precursors; (2) how the sensitivity changes as one moves from urban to regional scales; and (3) the relationship of  $O_3$  sensitivity changes to particle loading. The program we envision will include a network of comprehensive, chemical observing stations along a West-East transect extending from the Marin Headlands to Lake Tahoe, along the prevailing synoptic wind direction. The observing program is intimately integrated with a state-of-the-art coupled photochemical-meteorological model. Even the initial fraction of this capability that can be supported by LDRD would advance understanding of the chemical, meteorological, engineering, and public policy issues that are intertwined in the subject of urban, regional, and global pollution. To date, we have initiated construction of a second mobile laboratory and the instruments it would contain and have begun a comprehensive assessment of meteorological modeling appropriate for Central California.

## Accomplishments

We have integrated a modeling/measurement program to investigate air quality issues that are specific to California and to use our insights in these issues to illuminate our knowledge of air quality issues throughout the globe.

Specifically, we have built photochemical modeling capabilities for Central California, by assembling state-of-the-art descriptions of the atmospheric photochemical reaction mechanisms for VOC and  $NO_x$ , manmade and natural emissions of these pollutants, and prevailing meteorology for a period in summer 1998 when photochemical air pollution levels were high, and the measurement team was active at University of California (UC)'s Blodgett Forest, in California's Gold Country.

We implemented a new photochemical reaction mechanism based on work of Dr. William Carter. The mechanism was augmented to include reactions of methylbutenol, which is a hydrocarbon emitted by pine trees and has been a focus of experimental investigations at Blodgett Forest. We reviewed a series of air pollution episodes during summer 1998, and selected a period from August 26<sup>th</sup> through September 3<sup>rd</sup> when ozone levels were high on urban and regional scales. The period is of interest to regulatory agencies that we have built

relationships with, such as the Bay Area Air Quality Management District and the California Air Resources Board. The period in question also has good measurement coverage at the Blodgett Forest research site where measurements of  $NO_y$  and VOC were made. Revised estimates for isoprene emissions are greatly reduced, and lead to better agreement between measured and modeled isoprene and ozone.

An observing platform consisting of an integrated suite of state-of-the-art hydrocarbon, nitrogen oxide ozone, and aerosol measurements was assembled in a mobile trailer. The instrument suite was tested individually in the labs of Cohen and Goldstein and then used in a joint project at the edge of the Sacramento suburbs where we made measurements for six weeks during the summer of 2001. This experiment established a test of the framework for Lagrangian experiments linking observations at sites sequentially along the mean flow that was the basic design of this proposal. Ongoing analyses of the measurements in combination with measurements at the UC Blodgett Forest and in combination with the three-dimensional meteorological model are providing new insights.

## Publications

G.B. Dreyfus, G.W. Schade, and A.H. Goldstein, "Observational Constraints on the Contribution of Isoprene Oxidation to Ozone Production on the Western Slope of the Sierra Nevada, CA," submitted to *Journal of Geophysical Research*.

E.C. Wood, J.H. Freese, T.A. Albrecht, P.J. Wooldridge, and R.C. Cohen, "A Prototype Field Instrument for Detection of the Nitrate Radical by Laser-Induced Fluorescence," American Geophysical Union (AGU) Fall Meeting 2001 (November 2001).

M.B. Dillon, A.H. Goldstein, R.C. Cohen, *et al.*, "Chemical Evolution of the Sacramento Urban Plume: Transport and Oxidation," *Journal of Geophysical Research* (in press).

M.B. Dillon, A.H. Goldstein, R.C. Cohen, *et al.*, "On the Role of Nitrogen Oxides in the Sacramento Urban Plume," draft for *Journal of Geophysical Research*.

G.W. Schade and A.H. Goldstein, "Fluxes of Oxygenated Volatile Organic Compounds from a Ponderosa Pine Plantation," *Journal of Geophysical Research* **106**, 3111-3123, 2001 (November 2000).

G.W. Schade and A.H. Goldstein, "Plant Physiological Influences on the Fluxes of Oxygenated Volatile Organic Compounds from Ponderosa Pine Trees," *Journal of Geophysical Research* (in press).

G.W. Schade, G.B. Dreyfus, and A.H. Goldstein,  
"Atmospheric Methyl-*tertiary*-butyl-ether (MTBE) at a  
Rural Mountain Site in California" *Journal of  
Environmental Quality* (in press).

---

## **Indoor Bioaerosol Detection and Quantification by Polymerase Chain Reaction (PCR)**

---

Principal Investigators: William Fisk and Regine  
Goth-Goldstein

Project No.: 01020

### **Project Description**

Indoor airborne bioaerosols contribute to a broad range of health effects including communicable respiratory illnesses such as influenza and common colds. Measurements of the airborne concentrations and sizes of the virus or bacteria causing communicable respiratory illnesses are critical for research on the relationship of exposures to health effects, for studies of the influence of building characteristics on exposures, and for evaluations of exposure mitigation measures. However, measurement techniques have been especially limited for airborne virus, such as rhinovirus, which cause approximately half of common colds. Although controlled laboratory studies have indicated that rhinovirus is transmitted, in part, through the inhalation route, we have no detailed information on the size of aerosols with rhinovirus. We have only minimal information on the periods during illness when rhinovirus (RV) are shed, the airborne RV concentrations that cause illness, and the potential benefits of increasing building ventilation rates or filtration efficiency. Accordingly, the objectives of this project are to develop and demonstrate a quantitative polymerase chain reaction (PCR) assay for airborne rhinovirus in bioaerosols, as an alternative to current very onerous sampling and analysis methods, and to use this assay in controlled laboratory studies to learn about the size distribution of human-produced airborne particles containing rhinovirus.

### **Accomplishments**

#### *Development and Validation of Assay*

A quantitative PCR assay for human rhinovirus (RV) needs to be able to detect over 100 types of RV while distinguishing between the genetically similar rhinovirus

and enterovirus families. We used published genetic data to conduct a sequence analysis of RV and enterovirus families. Primers for the quantitative assay were chosen to target the 5' untranslated region of the RV genome, which has a high degree of genetic homology. With these primers, the assay generates a 203 base pair product that is specific for RV and will not detect the genetically similar enterovirus. The advantage of this assay is that no subtyping is needed to distinguish RV from enterovirus. This assay will detect more than 90% of RV strains. A few enterovirus-like RV strains (87, 14, 37, 72) are not detected.

RNA from RV type 89 was used to optimize the PCR reaction conditions in both a conventional thermal-block PCR cyclor and the rapid air microcapillary cyclor with real time product detection (LightCycler). A melting curve analysis of the PCR product in the LightCycler indicated that the product was pure. The PCR product of the LightCycler was examined by gel electrophoresis and was determined free of non-specific product. A quantitation standard for the PCR assay was obtained from RV type 89. Briefly, total RNA was isolated from RV 89 and reverse transcribed. The 425 base pair fragment that includes the amplification target was isolated and cloned into a bacterial cell line to produce as much of the quantitation standard as desired. The 425 base pair RV fragment was ligated into a pGEM vector. Competent *E. coli* cells (JM109) were transformed with the vector containing the RV fragment. Transformed cells were screened for the presence of the RV fragment by a simple PCR assay followed by a restriction enzyme digest. Many colonies had the RV quantitation standard. These colonies were grown in a larger culture; the vector containing the RV standard was isolated, purified, and quantitated by fluorescence.

Next, we examined the new RV PCR assay for a linear response. Using an eight-point serial dilution of the RV cDNA (spanning a 1000 fold concentration range), the LightCycler response was linear with a correlation coefficient ( $r^2$ ) of 1.00. The efficiency of the amplification reaction of the quantitation standard versus the viral RNA sample was virtually identical. The PCR products formed by the standard and the viral RNA sample were identical; thus, the internal standard will accurately reflect the presence of RV in samples.

The ability of the PCR assay to detect RV in individuals with a cold was tested. Seven volunteers were recruited and nasal lavage samples were tested for the presence of rhinovirus. Four of the six individuals with a cold tested positive for RV. The seventh individual, exhibiting allergy symptoms only, was negative for RV.



### *Collection of Aerosols with Defined Size Distribution and Detection of Rhinovirus in Particle Fractions*

The next stage of this project will quantify the size distribution of airborne RV-containing particles released by subjects with common colds. Volunteers reporting a common cold will work in a ventilated office-size chamber. Sampling of particles within the chamber in multiple particle size ranges and analyses to quantify RV will yield the size distribution. Airborne particles will be collected using a six-stage cascade impactor with size cuts of 7.1, 4.7, 3.3, 2.1, 1.1, and 0.65 microns plus a final filter with a 0.2-micron cut-off at the exit of the impactor. We have developed an isolation procedure to recover RV RNA from each stage of the impactor. To test the procedure, the RV RNA quantitation standard was spiked onto each stage of the impactor at varying amounts. After four hours of sampling, RNA was recovered with minimal degradation and successfully amplified with the RV PCR assay. Recovery varied from 80% to 50% depending on the initial amount spiked onto the plate. The procedure was able to recover and detect 1 nanogram of RV RNA. The stages were re-extracted to test the completeness of the isolation. At the 1-microgram level, only 1% of the initial RV RNA spiked onto the plate was recovered during the second extraction. At lower spiked amounts, no RV could be detected from the second extraction.

We recently performed an initial experiment with a volunteer who had a common cold. As it turned out, this volunteer's illness was not caused by a RV. Additional experiments will take place in the winter 2001-2002 cold season.

### **Publications**

M.L. Russell, R. Goldstein, M. Apte, and W. Fisk, "Size Distribution of Airborne Rhinovirus," abstract submitted, draft in preparation for Ninth International Conference on Indoor Air Quality and Climate, (June 30 – July 5, 2001).

---

### ***Diesel Particle Detection and Control***

---

Principal Investigators: Donald Lucas and Arlon Hunt

Project No.: 00013

### **Project Description**

Combustion is the primary source of fine particles (<2.5 microns) in the atmosphere. These particles are linked to

significant human health effects, but they are extremely difficult to measure and control. Conventional particle characterization techniques are inadequate to provide information at the rapid time scales necessary to study transient engine response (a major source of soot) or particle evolution as the exhaust is cooled and diluted. For this project, we utilized several low-cost sources for combustion particulates. We studied the characteristics of the particles emitted by these sources and how they evolved in the post-flame environment using two new instruments developed at Berkeley Lab. The particle studies were carried out using the diesel particle scatterometer (DPS) and eximer laser fragmentation fluorescence spectroscopy (ELFFS).

Past particulate research on diesels has focused on determining the development and role of the diesel exhaust particles in combustion. However, most prior research treats the particles only while they are in the combustion chamber. As soon as the exhaust is released from the combustion chamber, several mechanisms produce a rapid evolution of the particle characteristics. Cooling may cause vapors to condense into particles; these particles agglomerate and evolve rapidly in ways that depend very much on the details of the cooling and dilution conditions. It is these particles that are ultimately deposited into the environment and are cause for health concerns. This research extends ongoing work in the combustion region to the post-flame region, where the temperature, residence time, and concentrations of particles and other species are high. These conditions can foster extensive chemical and physical reactions between the components. Our goal is to understand these conditions to predict and control particulate and vapor components of diesel exhaust.

### **Accomplishments**

We utilized four different particle sources to study the evolution of particle characteristics between the source of production and their atmospheric release. These included a diesel and four-stroke gasoline generator, a two-stroke, spark-ignited engine, and an open flame system. In each case, the DPS and ELFFS and other instruments were used to examine the effect of varying the dilution and temperature history on the properties of the soot particles. In order to "freeze" the particle evolution at any given stage after combustion, an air injection pump was used to rapidly dilute and cool the exhaust gases after varying amounts of non-diluted cooling took place.

The DPS is a new instrument that provides a comprehensive analysis of three angle-dependent intensity and polarization transformations of light scattered from small particles. By fitting all three transformations simultaneously with the results of a scattering model, the DPS provides the size distribution and the real and

imaginary parts of the refractive index ( $n$ ,  $\kappa$ ) of the particles, thus providing some composition information. The DPS has a very rapid response, which was used to study the particle composition and size distribution during transient loading and unloading conditions as well as in the steady state. The DPS was used in conjunction with the ELFFS to study the dilution effects on particle size. Studies of the post combustion of diesel particles were also carried out using an afterburner/heat exchanger system. Results showed that the initial particle size of about 140 nanometers was reduced to less than 20 nanometers at the highest temperatures (800°C) and that the composition changed from an absorbing particle (elemental carbon) to a nonabsorbing component. This was interpreted as demonstrating the “burn out” of the carbon component of the particulate leaving a non-volatile nanoparticle component (probably sulfate). In a related analytical study, we were able to show that DPS can be used to correctly predict both the particle size and average real and imaginary parts of the refractive index for “porous” particles consisting of agglomerated primary diesel particles. It also predicted that the DPS will report the size of elliptical particles to be the mean between the major and minor dimensions.

ELFFS is an *in-situ*, atmospheric pressure technique that can measure a number of chemical species at elevated temperatures with good sensitivity, selectivity, and time response. It uses 193-nanometer photons from an ArF excimer laser to produce atoms or small fragments from larger compounds or particles that are normally non-fluorescing. The laser is moderately focused, but no breakdown or spark is produced from gas molecules. The ELFFS system was used to study the partition between hydrocarbon vapor and elemental carbon particles. The 248-nanometer fluorescent emission line from atomic carbon was used as a marker for soot particles. The hydrocarbon vapor concentration was determined from the CH and C<sub>2</sub> emission lines. The partition between these components was studied. By experiments using a denuder system to remove the vapor component from the exhaust stream, it was determined that the volatile component vanished, indicating that the adsorbed hydrocarbon species could be removed from the particles.

The results of this project established the general character of the particulate content of the post-combustion diesel exhaust; confirmed that the combustion products from diesel exhaust change rapidly with time, temperature, and dilution; determined that the exhaust contains both elemental carbon soot and volatile hydrocarbon species, and that there is an exchange between the vapor and particulate phase. In addition, we believe from our findings that the partition between vapor and solid phase can be controlled by post-combustion conditions. This provides a basis for optimizing the performance of particulate and vapor aftertreatment

devices by controlling the particulate component of the exhaust stream.

## **Publications**

P. Hull, I. Shepherd, and A. Hunt, “Modeling Light Scattering from Diesel Soot Particles,” submitted to *Applied Optics*.

## Life Sciences Division

---

### ***Quantitative Spatial and Temporal Resolution of Multicellular Interactions***

---

Principal Investigators: Mary Helen Barcellos-Hoff, Damir Sudar, Bahram Parvin, Abby Dernburg, Daniel Callahan, and Richard Schwarz

Project No.: 01021

#### **Project Description**

The purpose of this project is to provide an integrated resource for imaging of biological models in context. Quantitative imaging of complex biological materials is a critical area of the post-sequencing era. Current sequential measurements obtained with various microscopy techniques preclude detailed analysis of multidimensional responses. Quantitation of spatial and temporal concurrent behavior of multiple markers in large populations of multicellular aggregates is hampered by labor-intensive methods, a lack of quantitative tools, and the inability to index information. Ideally, one would track the kinetics and quantities of multiple target proteins, their cellular context, and morphological features in large populations. Future innovation is needed at the intersection of microscopy, all biology data acquisition strategies, image analysis, and high speed computing.

We will develop novel methodologies and propose that implementation of novel imaging algorithms—in conjunction with current Berkeley Lab capabilities in visual servoing, high-speed recording, and analysis—will allow the development of essential capabilities for simultaneous quantitative analysis of large multicellular populations. There are a number of examples of biological research at Berkeley Lab that require this technology. Such models and data will ultimately lead to the understanding of complex responses and accurate prediction of consequences in humans.

#### **Accomplishments**

In the first year of the project, the Life Sciences Division group is completing the optical microscopy instrument [three-dimensional-Visual Servoing Optical Microscopy (3D-VSOM) microscope] that will allow rapid and fully automated acquisition of dynamic three-dimensional images of live multi-cellular biological specimens and change the

experimental parameters under computer control. Simultaneously, the National Energy Research Scientific Computing Center (NERSC) group has extended their DeepView system with Declarative Flow Control facilities, which allow instantaneous re-configuration of the entire analysis software and network communication protocol to meet the requirements of specific experiments or unique features of the instrumentation controlled by the DeepView system. The DeepView system, combined with a predecessor of the 3D-VSOM system, was used to study multi-drug resistant human breast cancer cells. This has resulted in many optimizations to the DeepView environment and imaging instrument. A commercial restoration (deconvolution) microscopy system will be used to help characterize the performance of the 3D-VSOM system using standardized samples.

The 3D-VSOM system under development will be based on optical sectioning through the use of structured light rather than restoration through mathematical deconvolution. While the image acquisition for restoration microscopy itself is least photo-damaging for the live specimens, it was established in experiments on the microscopy system that a large number ( $> 30$ ) of serial images needs to be acquired to yield adequate data for the restoration procedure. So-called nearest-neighbor deblurring, which only requires three serial images, was considered inadequate based on preliminary experiments. Thus, this approach would cause a total acquisition time that is on the order of the biological processes that need to be monitored. Added to that, the significant computation time of the restoration procedure makes an experimental feedback loop impossible. Recent developments in the use of structured light to perform optical sectioning have resulted in a practical low-cost design, which was implemented for this project. A piezo-mounted Ronchi grating is located in the field diaphragm of the epi-fluorescence microscope and, from three images acquired with the grating period shifted by  $1/3$  each, a simple linear calculation yields an optically sectioned image with the out-of-focus information highly attenuated. It shares many of the benefits of restoration microscopy over confocal microscopy, such as low photo-damage or bleaching and white light excitation resulting in high flexibility in fluorochrome choice. Figure 23A shows the implementation of our design of the structured light-imaging microscope (SLIM), which was constructed in collaboration with David Wilson of the Berkeley Lab Engineering Division.

In the bioinformatic area, we have extended and implemented a new data model for storing dynamic information about how living cells are exposed to external compounds. As a result, measurement of uptake and

retention factor can now be linked with the compounds of interest. The bioinformatic framework, named BioSig, is distributed and allows clients to access their images along with the laboratory notebook data from their desktop. BioSig has been upgraded to accommodate three-dimensional datasets for subsequent retrieval and viewing. In the area of three-dimensional image analysis, we have extended our non-linear diffusion algorithm and Hough transform to process volume data. The techniques have been tested on synthetic three-dimensional images. The non-linear diffusion algorithm provides the basis of noise removal through harmonic analysis. The Hough transform is a generic tool for shape-based clustering using a predefined parameterized model. Automatic detection of nuclei from volumetric dataset is an important step in large-scale analysis of scientific microscopy data. These datasets may represent organization of nuclei in a cultured colony, and successful detection of each nucleus and its relationship to the organization of the colony provides the basis for better understanding of cell-cell communication under a variety of treatments and conditioning. An efficient algorithm has been developed to detect nuclei of interest in three-dimensions. In general, detection of individual nucleus is a complex process due to the fact that nuclei of interest:

- Do not respond uniformly to the fluorescent compounds.
- May have many internal substructures.
- Overlap each other as a result of cell division.

The first step of our protocol is scale space extraction and classification of elliptic features. The classification result is then used to group similar features using a three-dimensional connected component algorithm. Although false labeling is unavoidable, sufficient information can be gathered so that a higher-level constraint can group partial information into a whole. This higher-level constraint is expressed in terms of convexity and implemented as a convex hull for improved efficiency. The proposed framework has been applied to a collection of multicellular systems for detailed quantitative analysis, and the result is shown in Figure 23B.

The ability of Berkeley Lab investigators to contribute and compete in biology is augmented by state-of-the-art imaging strategies.

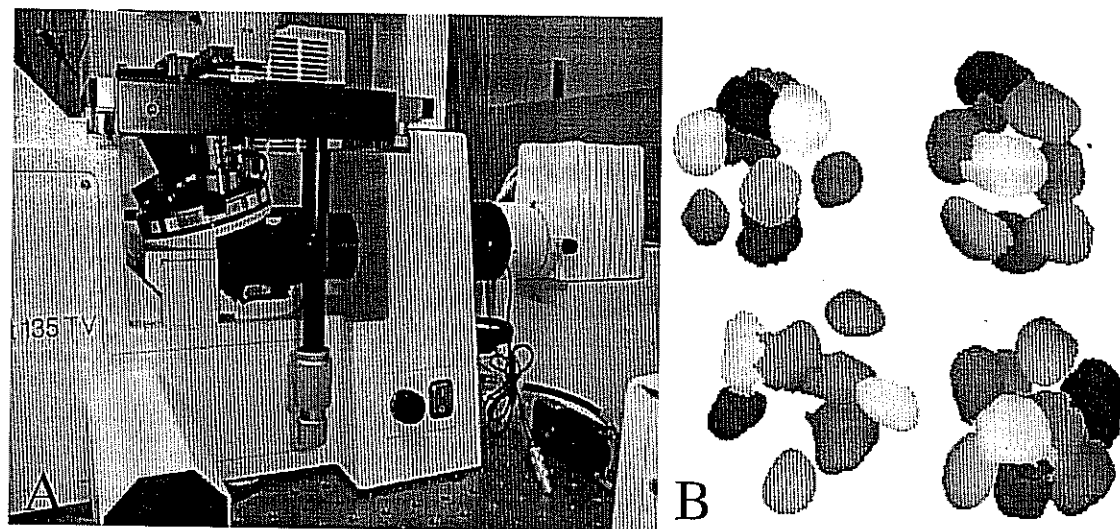


Figure 23: Panel A shows the Berkeley Lab SLIM microscope; panel B shows automatically segmented three-dimensional images of mammary cell cultures.

## Publications

C. Ortiz de Solorzano, S. Costes, D.E. Callahan, B. Parvin, and M.H. Barcellos-Hoff, "Applications of Quantitative

Digital Image Analysis to Breast Cancer Research," in press for *Microscopy Research and Techniques*.

D. Sudar, D. Callahan, B. Parvin, D. Knowles, C. Ortiz de Solorzano, and M.H. Barcellos-Hoff, "Design of a Microscopy System for Quantitative Spatial and Temporal Analysis of Multicellular Interactions," *Proceedings of*

*Microscopy and Microanalysis 2001 Meeting* (August 5-9, 2001).

D. Sudar, B. Parvin, D.E. Callahan, R.I. Schwarz, D.W. Knowles, C. Ortiz de Solorzano, and M.H. Barcellos-Hoff, "Microscopy Environment for Quantitative Spatial and Temporal Analysis of Multicellular Interactions," draft for *Proceedings of Photonics West – BiOS Biomedical Optics 2002 Conference* (January 19-25, 2002).

B. Parvin, Q. Yang, G. Fontenay, M.H. Barcellos-Hoff, "BioSig: An Imaging Bioinformatic System for Studying Phenomics," submitted to *Institute of Electrical and Electronics Engineers Conference Computer*.

---

## **Positron Emission Tomography (PET) Ligands for the N-methyl-D-aspartate (NMDA) Receptor Channel**

---

Principal Investigators: Anat Biegon

Project No.: 01022

### **Project Description**

The ultimate purpose of this project is to develop and validate a new brain imaging agent which would be used to optimize and monitor treatment of stroke and brain injury and to provide new insights into the etiology and treatment of addictive behavior and other brain disorders linked to increased or decreased activity of glutamate, the major excitatory neurotransmitter in the brain. The immediate goal is to synthesize, screen, and optimize new radiopharmaceuticals suitable for *in-vivo*, non-invasive imaging of activated brain glutamate NMDA receptors by positron emission tomography (PET).

New molecules suitable for labeling with positron emitters (Fluorine-18, Carbon-11) will be synthesized from lead compounds and screened by *in-vitro* binding methods for improved affinity and lipophilicity. Specificity to the NMDA receptor will be assessed by a combination of *in-vitro* binding and autoradiographic techniques. *In-vivo* selectivity and kinetics will be evaluated by autoradiography and biodistribution studies. The best molecule will be labeled with Carbon-11 or Fluorine-18, and investigated in dogs for biodistribution, dosimetry, and safety. Two-species subacute toxicology will be evaluated with the "cold" compound if necessary, in preparation for evaluation in humans.

### **Accomplishments**

#### ***Chemical Synthesis and In-Vitro Screening of New Fluorine-Containing Derivatives Cerestat***

Five new molecules were synthesized and purified. All were full antagonists at the receptors with affinities ranging from 5.5 to 110 nanomolar (nM). All had logP (P = octanol/water partition coefficient, the higher the number the more lipophilic the compound values in the target range ( $1 < \log P < 2$ )).

#### ***Tritium Labeling of CNS-5161***

Dr. Philip Williams and Hiromi Morimoto of the National Tritium Labeling Facility, with the help of Dr. Gibbs, have devised the route for tritiating CNS-5161, the cerestat analog with the highest affinity we found so far. Adapting the synthetic strategy to allow for chemistry on a micromole ( $\mu\text{M}$ ) scale and to reduce the use of chlorinated solvents, tritiated CNS-5161 was successfully prepared. The first batch (May 2001) resulted in 96 millicuries of tritiated material with a final specific activity of 66 Ci/mmol.

#### ***In-Vitro and In-Vivo Characterization of [ $^3\text{H}$ ]CNS-5161***

The association and dissociation kinetics, affinity, and pharmacological specificity of [ $^3\text{H}$ ]CNS-5161 were evaluated in rat forebrain homogenates.

*Association and dissociation kinetics* were determined at room temperature as well as at 37°C, to get a closer approximation for the likely *in vivo* kinetics of the candidate agent. We found that time to equilibrium was 90 minutes at room temperature and 30 minutes at 37°C. Dissociation was induced by addition of 10  $\mu\text{M}$  MK801 and the binding was fully reversible with a half-life of 20 minutes.

*Saturation characteristics*: The dissociation constant ( $K_d$ , a measure of affinity to the receptor) and the maximal number of binding sites ( $B_{\text{max}}$ ) for CNS-5161 were calculated through Scatchard analysis. The binding was saturable within the concentration range used with a linear scatchard plot, suggesting a single binding site. Mean  $K_d$  was 2.3 nM (twice the affinity of the gold standard MK801).

*Glutamate/glycine dependence*; a hallmark of noncompetitive, use-dependent NMDA antagonists, was examined by running parallel saturation experiments in the absence and presence of excess glutamate and glycine. Specific binding was reduced by more than 50% in the absence of exogenously added glutamate and glycine.

*Pharmacological specificity*: The only compounds that inhibited binding of [ $^3\text{H}$ ]CNS-5161 were the NMDA antagonists. Drugs acting on other brain receptors and transporters (e.g. adrenergic, serotonergic, sigma, cannabinoid, opioid and dopamine markers) had no effect

on the binding even at very high (10 $\mu$ M) concentrations. Studies of the biodistribution, brain kinetics and specificity of [<sup>3</sup>H]CNS-5161 *in vivo* have been initiated. CNS-5161 was found to have adequate brain penetration [maximal percent of injected dose (%ID) found in the brain = 1.47]. It is taken up avidly by liver and lung. Untreated and NMDA-treated animals demonstrate specific (i.e. MK801-displaceable and regionally selective) binding *in-vivo*.

## Publications

A.R. Gibbs, H. Morimoto, H.F. VanBrocklin, P.G. Williams, A. Biegon, "Synthesis of N-(2-chloro-5-methylthiophenyl)-N'-(3-methyl-thiophenyl)-N'-[<sup>3</sup>H]methylguanidine, {[<sup>3</sup>H]CNS-5161}," accepted for publication in *Journal of Labelled Compounds and Radiopharmaceuticals*, (October 2001).

A. Biegon, A. Gibbs, M. Ono, M. Alvarado, and H.F. VanBrocklin, "Synthesis and *In-Vitro* Evaluation of Novel Brain NMDA Receptor Channel Imaging Agents," *Proceedings of Society for Neuroscience Meeting* (November 2000).

M. Ono and A. Biegon, "A Novel Tritiated Non-Competitive NMDA Antagonist [<sup>3</sup>H]CNS-5161: *In-vitro* Characterization," (in preparation).

A. Biegon, M. Alvarado, M. Ono, S.E. Taylor, and H.F. VanBrocklin, "A Novel Tritiated Non-Competitive NMDA Antagonist [<sup>3</sup>H]CNS-5161: *In-vivo* Characterization," (in preparation).

---

## A High Sensitivity In-Vivo Crosslinking Method

---

Principal Investigators: Mark Biggin

Project No.: 01023

## Project Description

Metazoans are distinguished from microbes by their extraordinarily complex arrangements of cells. The differences between cells within an animal are fundamentally differences in gene expression; and one of the most prominent levels at which gene activity is regulated is at the level of RNA transcription. Sequence-specific DNA binding proteins, any one of which can bind tightly to only a limited number of DNA sequences, are the principle agents used to selectively control gene transcription. Our long-term goal is to characterize the

range of DNA sequences bound by transcription factors in animals, the forces that determine the range of DNA sequences bound, and the function of factors bound to their targets. This goal is challenging for many reasons: for example, animal regulatory networks are vastly more complex than those of microbes; metazoan promoters and genomes are much longer; and many metazoan factors recognize shorter, more degenerate DNA sequences. All of which makes it impossible to predict the *in-vivo* pattern of DNA binding from simple measurements of *in-vitro* DNA affinities.

To overcome the above challenge, we previously developed a method that directly measures relative occupancy of specific factors on DNA in intact, living animals. We have examined *in-vivo* binding of the homeoprotein family of developmental regulators in the fruit fly *Drosophila melanogaster* as this is probably the most experimentally tractable system available: *Drosophila* is an outstanding model organism for genetic and transgenic analysis; the developmental regulatory cascade initiating development is best understood in this organism; and the complete genome sequence is available—allowing system-wide studies to be conducted. Our results indicate that the pattern of homeoprotein DNA binding differs dramatically from that predicted by indirect experiments. These proteins bind in a surprising manner to sites throughout the length of most genes. Further, complementary genetic experiments indicate that each homeoprotein regulates the expression of a majority of genes; and a variety of molecular data imply that most of these genes are directly controlled. Our work aims to better characterize the above observations and to determine how transcription factors that bind so broadly can coordinate complex and precise events such as morphogenesis.

There are two limitations of our current *in-vivo* DNA binding data. First, we can only measure interactions to 0.5 to 2 kb regions of DNA that contain 5 to 15 recognition sites for a specific factor. Thus, we cannot be certain which particular elements are occupied, hampering our ability to determine the functional significance of the widespread DNA binding we observe. Second, we can only measure binding to one DNA fragment at a time. It would be much better if we could look at binding to thousands of DNA regions simultaneously. This project seeks to overcome these two problems.

## Accomplishments

Progress on this first limitation, higher resolution mapping, has been slow as it has taken till now to have our fly incubator built. This is about to happen and the purchases of this room have absorbed most of the first year costs.

We have made great strides on addressing the second limitation. We have set up a genomic microarray *in-vivo*

crosslinking method that can measure binding to thousands of one kb genomic fragments at one time. This work has been done in collaboration with the Berkeley *Drosophila* Genome Project as part of a large multi-laboratory collaboration that the Principal Investigator is organizing to study the transcriptional network in the early *Drosophila* embryo. Using crosslinked chromatin derived from tissue culture cells, we have established a basic protocol that works. The sensitivity of the assay is currently not as high as our original southern blot method, but recently we have shown that the microarray method is quantitative. We are about to use the microarray method to look at binding in embryos, rather than tissue culture cells, and will continue to optimize the sensitivity of our protocol. These experiments should be completed soon.

---

## **Transgenic Mouse Models for DNA Damage Sensing, Repair, and Aging**

---

Principal Investigators: David Chen

Project No.: 00015

### **Project Description**

The purpose of this proposal is to understand the role of DNA repair proteins in cellular senescence and telomere maintenance in mammals. We hypothesize that DNA double-strand break (DSB) repair proteins are involved in telomere metabolism at the end of chromosomes. Cellular deficiency in expression of DSB repair proteins would lose the ability to protect the chromosome ends. We propose to construct knockout transgenic mice deficient in DNA double-strand break repair and DNA base damage repair. Cellular senescence in primary fibroblasts and telomere length maintenance in embryonic stem (ES) cells will be determined. The specific aims are:

- To generate knockout transgenic mice involved in DSB repair.
- To determine the telomere maintenance and cellular senescence in mouse embryonic fibroblasts (MEFs) isolated from transgenic mice deficient in Ku70, Ku80, DNA-dependent protein kinases (DNA-PKcs) and the double knockout Ku80/DNA-PKcs.

We will test whether deficiency in DNA double-strand repair proteins in mammalian cells would accelerate telomere shortening, thereby resulting in premature cellular senescence. We will analyze primary fibroblasts from our

existing knockout mice, Ku70, Ku80, DNA-PKcs, and the double knockout Ku80/DNA-PKcs to determine their possible role in cellular senescence.

### **Accomplishments**

#### ***Disruption of NBS1 Gene Leads to Early Embryonic Lethality in Homozygous Null Mice and Induces Specific Cancer in Heterozygous Mice***

Nijmegen breakage syndrome (NBS) is a rare autosomal recessive chromosome instability syndrome characterized by microcephaly, growth retardation, immunodeficiency, and cancer predisposition. Cells from NBS patients are hypersensitive to ionizing radiation with cellular features indistinguishable from ataxia telangiectasia (AT). However, NBS is genetically distinct from AT and has been shown to result from mutations in a novel mammalian gene *Nbs1* that codes for a 95-kilodalton protein called nibrin, NBS1, or p95. NBS1 is part of the Mre11-Rad50 complex that becomes associated with DNA DSBs in irradiated cells early in the cellular DNA-damage response indicating that it may function in the early stages of detection and signaling of DNA damage.

To establish an animal model for NBS, we initiated the process of constructing NBS1 knockout mice. However, NBS1 gene knockouts were embryonic lethal at an early embryonic stage. NBS1 homozygous(-/-) blastocyst cells cultured in vitro showed retarded growth and subsequently arrested their growth within five days. Prominent apoptotic cell death was observed in four days in cultured NBS1 homozygous(-/-) blastocyst cells by TUNEL assay. NBS1 heterozygous(+/-) mice are normal, and exhibit no specific phenotype for at least up to one year. However, fibroblast cells from NBS1 heterozygous(+/-) mice showed enhanced transformation compared to NBS1 wild-type(+/+) cells. Also the heterozygous mice showed high incidence of hepatocellular carcinoma after reaching one-year-old, compared to wild-type mice, even though there was no critical difference of other tumor incidence, such as lung adenocarcinoma and lymphoma. This strongly suggests that NBS1 heterozygosity and reduced NBS1 expression induce a specific tumor in mice.

#### ***Construction of Conditional NBS Knockout Mice***

In order to avoid the lethality associated with NBS gene knockout, we worked on establishing conditional knockouts for NBS using the tetracycline (tet) inducible system (Tet-On system, available from Clontech). Also, viable mice which escape from embryonic lethality and whose NBS gene expression can be turned off after birth would be a good model for human NBS patients and would be useful for analyzing basic functions of NBS protein *in vivo*.

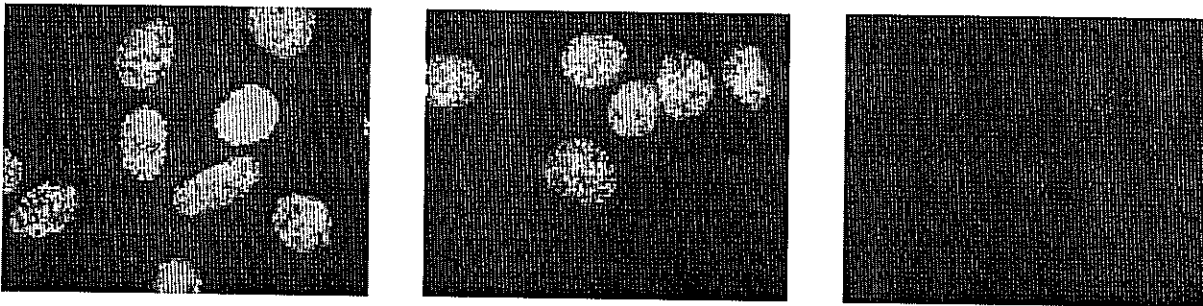
Using a newly designed Tet-Off/CreLoxP conditional knockout approach, we have obtained three independently

targeted embryonic stem (ES) cell clones. From these three ES cell clones, we have generated 16 chimeric mice whose chimerism range from 20% to greater than 90%. We do not expect serious obstacles in obtaining the heterozygote and homozygote mice since several mice show a high percentage of chimerism (>90%). In addition, we have previously used the same ES cells to successfully generate DNA-PKcs knockout mice successfully. Since August 2001, we have started to cross these chimeric mice with C57BL in order to generate Nbs<sup>±</sup> heterozygous mice. We have already obtained two mice that are heterozygous for the Nbs1 conditional knock out gene. These mice were derived from chimeric mice generated from ES line 25D. All 12 pups from the birth show agouti color and two out of five pups examined were positive for the conditional Nbs1 knock out gene as confirmed by polymerase chain reaction (PCR)-based genotyping. Southern blot analysis will also be carried out to further confirm the heterozygosity. Thus far, chimeric mice derived from 1F and 25A ES lines have not produced positive heterozygous mice. Currently, we are continuously breeding the 25D line, as well as 1F and 25A lines to obtain more F1 heterozygous mice. Apart from the

two heterozygous mice already obtained, we expect to obtain more heterozygotes from the mice currently being mated. We expect that we will start to cross the F1 heterozygous mice to create homozygous mice for analysis of NBS functions.

#### *Phosphorylation of Histone H2AX by Ataxia Telangiectasia-Mutated (ATM) is "Backed Up" by DNA-PK:*

We have in the last few months completed a new project aimed at the identification of the kinase responsible for histone H2AX phosphorylation upon DNA damage. Our results, identifying ATM as the major kinase responsible for H2AX phosphorylation, has been recently published by *Journal of Biological Chemistry*. We show in the Figure, a comparison of H2AX focus-formation between wild type, DNA-PKcs<sup>-/-</sup>, and Atm<sup>-/-</sup> mice. An interesting concept that emerged from our study was that DNA-PK could serve as a "back-up" kinase for ATM in this regard which supports our overall hypothesis of cross-talk between DNA-PK and ATM.



*Figure 24: Immunostaining of irradiated wild type, DNA-PKcs<sup>-/-</sup>, Atm<sup>-/-</sup> cells with anti-phospho-H2AX antibody. Low levels of immunostaining in Atm<sup>-/-</sup> cells is due to "back-up" phosphorylation by DNA-PK.*

#### **Publications**

A. Kurimasa, S. Burma, M. Henrie, H. Ouyang, M. Osaki, H. Ito, H. Nagasawa, J.B. Little, M. Oshimura, G.C. Li, and D.J. Chen, "Disruption of Nbs1 Gene Leads to Early Embryonic Lethality in Homozygous Null Mice and Induces Specific Cancer in Heterozygous Mice," (in preparation).

S. Burma, A. Kurimasa, R.B. Cary, H.A. Crissman, G. Iliakis, N.D. Lakin, G.C.M. Smith, G.C. Li, S.P. Jackson, and D.J. Chen, "Persistent ATM Activation in Ku80<sup>-/-</sup> Fibroblasts: A Potential Role for Ku as a Modulator of ATM Kinase Activity," (in preparation).

S. Burma, B.P. Chen, M. Murphy, A. Kurimasa, and D.J. Chen, "ATM Phosphorylates Histone H2AX in Response to DNA Double-Strand Breaks," *Journal of Biological Chemistry* (2001).



---

## Dynamic Reorganization of Chromosome Architecture during Meiosis

---

Principal Investigators: Abby Dernburg

Project No.: 01024

### Project Description

All sexually reproducing organisms rely on a special cell division process called meiosis to generate gametes, which contain only a haploid number of chromosomes. The central event of meiosis is a reductional division in which pairs of homologous chromosomes must segregate to different daughter nuclei. The accuracy of this unique division depends on recombination, the presumed *raison d'être* of the entire process, which in turn requires that each chromosome must find and pair with its appropriate homologous partner. Errors in meiosis lead to missegregation, with disastrous consequences if an embryo inherits an abnormal number of chromosomes.

Meiosis presents a dramatic example of large-scale nuclear reorganization in response to a specific differentiation program. Many aspects of this process are still mysterious, with no clear precedent in our molecular understanding of biology. How do chromosomes first encounter their partners? How does each recognize its homolog? How do they achieve side-by-side alignment along their entire lengths? How does the protein scaffold called the synaptonemal complex assemble between partner chromosomes? Within this structural context, what regulatory mechanisms ensure that each chromosome undergoes reciprocal recombination while placing tight restrictions on the number and placement of crossovers? How are these nuclear events coordinated with the developmental program giving rise to sperm, oocytes, spores, or pollen? The mechanisms used by the meiotic cell to orchestrate all of these complex processes are virtually unknown.

We will study the complex macromolecular structures that are built and disassembled during meiosis. We will harness both the molecular genetic and cytological advantages of the tiny soil-swelling nematode *C. elegans*. This integrated approach will bridge the gap between individual gene products and large-scale meiotic structures.

The long-term goal of this project will be to understand the relationship between structural elements and their function in the meiotic nucleus. In our initial experiments we will:

- Visualize and quantify the dynamic properties of meiotic chromosomes.
- Probe the role of chromosome architecture in controlling the distribution of crossovers.
- Investigate the essential role of the meiotic "pairing center" and associated proteins.

The project will integrate several technological approaches, including three-dimensional imaging, functional genomics, and genetics. We will use genetics to identify and characterize novel components of the chromosome-organizational machinery that remodels the nucleus during meiosis. Using high-resolution fluorescence imaging, we will localize molecular components at the subcellular level to learn how they contribute to nuclear modeling.

### Accomplishments

The major equipment purchase made from these funds was an optical-sectioning microscope and image deconvolution software. This system represents the state-of-the-art in high-resolution, three-dimensional microscopy, and we are now using it extensively. A significant portion of the FY 2001 effort was in establishing this experimental set up.

A new postdoctoral fellow, Peter Carlton, began working in the lab in August. He is primarily working toward the first goal of this Laboratory Directed Research and Development (LDRD) project, the visualization and quantitation of meiotic chromosome dynamics. So far, he has achieved two significant technical advances: (1) he has identified a suitable method for mounting individual worms on microscope slides to facilitate *in-vivo* imaging (this has been a challenge because worms tend to crawl around unless they are anaesthetized with drugs, which we prefer to avoid using since they may have deleterious effects on chromosome structure and function) and (2) he has written a simple but clever computer algorithm that allows him to align both individual optical sections and successive time points in a series of images. This is critical because our image processing and analysis methods require that the images have minimal offset with respect to each other. Since there is considerable movement of meiotic nuclei within living animals, this has been an important step in enabling us to image chromosome movement within the nuclei.

Collaborations with several other investigators have also been initiated. For example, I am working with Chang-Su Lim and Judy Campisi [(Life Sciences Division (LSD)] to

characterize the role of a telomere-regulatory protein, CLK-2, which they had identified in *C. elegans*. Based on the cytological phenotype we observed in *clk-2* mutant worms, a likely role for this protein was suggested in mediating a DNA damage checkpoint response, a hypothesis that was eventually supported by our work and that of other laboratories. In addition, I provided microscopy assistance to the laboratory of Terumi Kohwi-Shigematsu, who has submitted a manuscript that includes this work. Towards our future goals, I have established a collaboration with Bahram Parvin [National Energy Research Scientific Computing Center (NERSC)], which we hope will enable us to meet new technical challenges in analyzing complex real-time images. We are also currently attempting to apply novel algorithms developed by Stephen Holbrook (Physical Biosciences) and Inna Dubchak (LSD, NERSC) to discover functional noncoding RNA molecules (fnRNAs) in the *C. elegans* genome, which potentially would have enormous impact on our investigations. Through funding provided by a second LDRD, we are also working with engineers Tony Hansen and Jian Jin to develop new genetic transformation technology for *C. elegans*, which (if successful) will greatly augment our ability to pursue the goals of this project.

## Publications

A.F. Dernburg, "Here, There, and Everywhere: Kinetochore Function on Holocentric Chromosomes," *Journal of Cell Biology*, 153, S33-8 (June 11, 2001)  
<http://www.jcb.org/cgi/content/full/153/6/F33>.

C.-S. Lim, S. Mian, A. Dernburg, and J. Campisi, "*C. elegans clk-2*, a Gene that Limits Life Span, Encodes a Regulator of Telomere Metabolism Similar to Yeast Telomere Binding Protein Tel2p," in press for *Current Biology*.

---

## Experimental and Computational Analysis of the Mechanisms and Logic of Eukaryotic Transcriptional Regulation

---

Principal Investigators: Michael Eisen

Project No.: 00016

## Project Description

The focus of our work is on characterization of the mechanisms and logic of transcriptional regulation in the

yeast *Saccharomyces cerevisiae*, and, more recently, in the fruit fly *Drosophila melanogaster*. In yeast, we are particularly interested in understanding how variation in gene expression contributes to variation in phenotype, and in understanding how sequence variation in non-coding regions of the genome generate variation in gene expression. We employ a combination of experimental and computational approaches to address these questions. Our experimental work utilizes whole-genome DNA microarrays to monitor the expression of every gene in the genome. We use these arrays to monitor gene expression changes following environmental perturbations (changes in temperature, nutrient availability and the concentration of toxic or potentially toxic metabolic byproducts such as ethanol). We are currently performing identical experiments in yeast strains with different genetic backgrounds (we work with a variety of laboratory strains and commercial and naturally isolated wine yeasts) and are characterizing differences in the environmental responses of the different strains.

The goal of these studies is to relate the observed differences in gene expression responses to differences in the cells' phenotypic response to the treatments. We hope both to understand how significant a role transcriptional regulation plays in phenotypic variation, and to understand the relationship between specific gene expression changes and specific phenotypic responses. In parallel, we are developing computational methods to explore how transcriptional information is encoded in the yeast genome sequence, to build predictive models that relate genome sequences to gene expression patterns, and to understand how variation in genome sequences will alter gene expression responses. Finally, we are beginning a project that combines phenotypic analysis, gene expression analysis and whole-genome genetic mapping techniques to directly link complex organismal phenotypes to gene expression and genome sequence.

We believe the method we are using will be a prototype for understanding the genetic basis of complex traits in more complex systems such as humans. In *Drosophila*, we are also interested in understanding how transcriptional regulatory information is encoded in the (fully-sequenced) *Drosophila* genome, with a focus on the transcriptional regulatory cascades that choreograph the complex developmental events that specify the form and function of the adult fly. Unlike in yeast, where transcriptional information for a given gene is contained in a relatively small piece of DNA immediately upstream of the gene, in animals like *Drosophila*, transcriptional information for a single gene is often spread over tens of thousands of basepairs. Therefore, the proximal challenge in analyzing the *Drosophila* (and also human and mouse) genomes is in identifying the regions of the genome that contain transcriptional information. We have begun to develop and

apply novel computational methods to identify such regions.

## Accomplishments

### *Experimental Analysis of Gene Expression in Yeast*

We are now routinely printing whole-genome yeast microarrays in the lab. These arrays consist of single 70 basepair synthetic oligonucleotides for each gene. We have refined experimental conditions and now are using these arrays to study gene expression variation in a series of different yeast strains. We have examined gene expression responses to changing temperature, starvation, diauxia (the shift from fermentable to non-fermentable carbon sources), anoxia, and variable ethanol concentrations in four standard strains (two lab strains, a commercial wine strain and a field isolate). Much of the response is conserved amongst these phenotypically distinct strains, but there are striking differences that are likely to be directly related to the phenotypic differences. We are currently analyzing these relationships in more detail.

### *Computational Analysis of Transcriptional Regulation in Yeast*

We developed a new method for analyzing the relationship between the yeast genome sequence and large volumes of gene expression data. All previously published methods took an approach that relied first on the analysis of expression data to identify genes with similar patterns of expression, followed by an analysis of the genome sequence to search for sequence features shared by coexpressed genes. Our approach—the computation of genome-mean expression profiles (GMEPs)—takes the opposite approach and systematically examines all possible sequence features (in some family, such as all 6mers) and evaluates the transcriptional information each contains. We successfully applied GMEPs to a number of different datasets, recovering many known transcription factor binding sites and identifying a number of novel sites. The method was presented at the International Conference on Intelligent Systems in Molecular Biology and published in *Bioinformatics*. Our software implementation of the GMEP algorithm has been downloaded over 500 times in the last six months. We are currently extending the algorithm in a variety of ways.

In collaboration with groups in the University of California, Berkeley Biostatistics and Statistics Departments, we have been exploring and developing methods to build models that would allow us to accurately predict all genes'

expression patterns on the basis of the genome sequence. Although there is a long way to go before we reach this goal, in the six months we have been working on this project, we have developed a number of different algorithms, based on classification trees, multivariate regression and some novel methods that show great promise. For example, there are approximately 100 genes in the genome that contain a consensus-binding site for the transcription factor PHO4. However, only 20 of these genes actually respond to altered phosphate levels (the trigger for activating PHO4). A classification tree based on motif positioning and DNA structural properties was able to distinctly identify 60 of the 80 non-responding genes, providing a remarkable increase in our ability to predict which genes will show a transcriptional response to changing phosphate levels.

Finally, we have developed and exhaustively evaluated a number of non-deterministic (fuzzy) clustering methods to augment standard methods used for the analysis of microarray data. These methods are far more compatible with the realities of transcriptional regulation, in which many genes are regulated by multiple regulatory pathways and can thus have transcription patterns that are biologically significantly similar to those of many distinct transcriptional families.

### *Computational Analysis of Transcriptional Information in the Drosophila Genome*

Just as coding genes in animals are organized in separate modular units (exons), transcriptional information in animals is also modularly organized. We have recently developed a computational method to identify cis-regulatory modules (CRMs) in the *Drosophila* genome based on the density of predicted binding sites for transcription factors that are known to act in a combinatorial manner. When we applied this method to find CRMs responsive to transcription factors active in the early *Drosophila* embryo, we were able to successfully recover 14 of 19 known CRMs, and identified at least 30 new binding site clusters that are potentially unrecognized CRMs. A strongly statistically significant number of these novel clusters are adjacent to genes that have patterned expression in the early embryo. We tested one of these novel clusters experimentally and confirmed that it is, indeed, a CRM that controls the expression of the adjacent gene. The method and these results will be appearing early in 2002 in *Proceedings of the National Academy of Sciences*.

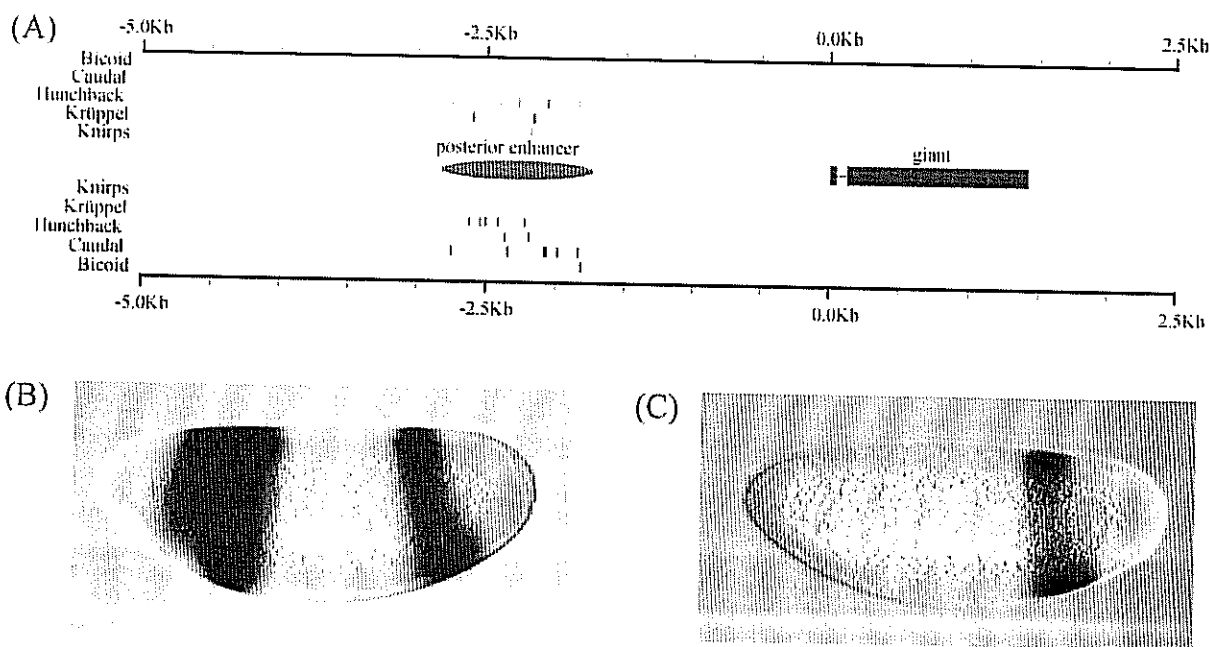


Figure 25: (A) Cluster of binding sites found between 2.9 kilobases and 1.8 kilobases upstream of *giant*. The DNA segment surrounding the cluster (labeled "posterior enhancer") was cloned into a *lacZ* fusion construct and introduced into the genome via germline transformation. (B) Normal expression of *giant* in syncytial blastoderm embryos. (C) Expression of the predicted CRM-*lacZ* transgene in an embryo at the same stage.

## Publications

D.Y. Chiang, P.O. Brown, and M.B. Eisen, "Visualizing Association Between Genome Sequences and Gene Expression Data Using Genome-Mean Expression Profiles," proceedings of International Meeting on Intelligent Systems in Molecular Biology published in *Bioinformatics*, 17, S49-55 (June 2001)  
[http://rana.lbl.gov/papers/Chiang\\_ISMB\\_2001.pdf](http://rana.lbl.gov/papers/Chiang_ISMB_2001.pdf).

B.P. Berman, Y. Nibu, B. Pfeiffer, P. Tomancak, S. Celniker, M.S. Levine, G.M. Rubin, and M.B. Eisen, "Exploiting Transcription Factor Binding Site Clustering to Identify Cis-Regulatory Modules Involved in Pattern Formation in the *Drosophila* Genome," in press for Proceedings of the National Academy of Sciences (January 2002).

A.P. Gasch and M.B. Eisen, "Fuzzy Clustering Analysis of Gene Expression," (in preparation).

S. Keles, M. van der Laan, and M.B. Eisen, "Identification of Regulatory Elements Using a Feature Selection Algorithm," (in review).

## Tracking Proteins in Light and Soft X-Ray Microscopy

Principal Investigators: Carolyn Larabell

Project No.: 01025

## Project Description

X-ray microscopy can be used to localize proteins in whole, hydrated cells at 5 to 8 times better resolution than possible with light microscopy. Until recently, however, the information was presented as a two-dimensional projection of three-dimensional data. We are currently developing the technology to do tomographic reconstructions of whole cells using x-ray tomography. We will use light microscopy to examine the dynamics of specific proteins that have been tagged with the fluorescent marker called green fluorescent protein (GFP) in live cells. After the dynamics of the protein have been determined, the cells will be fixed, labeled with commercially available antibodies to GFP, and then examined in the x-ray microscope to determine the

subcellular location of the protein. The use of the x-ray cryo-tomography will generate unique, high-resolution three-dimensional information about the subcellular localization of the protein throughout the entire cell. The development of this methodology will provide a high-throughput method for structure-function analyses of proteins.

Our initial studies will be designed to demonstrate the feasibility of these studies. We will transfect human mammary epithelial cells with GFP constructs for proteins with characteristic labeling patterns. The dynamics of the fluorescent proteins will be examined by monitoring the living cells using confocal or multi-photon microscopy. Optical sections of the cells will be collected over time and three-dimensional reconstructions of the labeled proteins within the cell will be obtained. After the live cell studies are completed, the cells will be fixed and labeled using commercially available anti-GFP antibodies followed by immunogold labeling for the x-ray microscope. A series of images at 150 different angles will be collected for tomographic reconstruction.

## Accomplishments

The research proposed requires several technological developments. We have made progress in two main areas, as described below.

### *Tracking Green Fluorescent Protein (GFP)-Tagged Proteins*

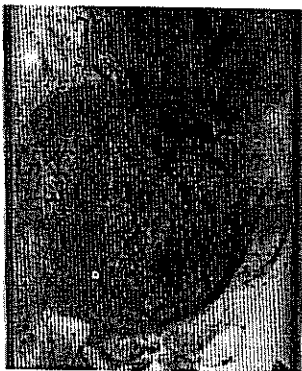
We have demonstrated the ability to track protein constructs tagged with the green fluorescent protein (GFP) in living cells using standard laser scanning confocal microscopy as well as multi-photon microscopy. In some cases, mRNAs for the proteins were injected into *Xenopus* embryos at the two-cell stage (e.g. mRNA tagged with GFP encoding the protein known as Tumorhead or the transcription factor, Xnf-7, in collaboration with Dr. Laurence Etkin from M.D. Anderson Cancer Center; and mRNA tagged with GFP encoding Protein Kinase C). These mRNAs were translated into proteins and those fluorescently tagged proteins were examined in later developmental stages using time-lapse imaging techniques. These experiments reveal that each of these proteins moves around the cell throughout the cell cycle, residing at the cell surface, in the cytoplasm, or in the nucleus. The mechanism(s) by which they are transported around the cell or by which they are sequestered at each of these sites remains unclear. Elucidation of these mechanisms will be aided by follow-up studies using high-resolution soft x-ray microscopy as described below.

For other experiments, DNAs encoding the proteins (beta-catenin, actin, and Protein Kinase C) were inserted into human mammary epithelial cells via electroporation. These cells were monitored at various times between four

hours and five days and the GFP protein constructs were tracked using time-lapse imaging techniques. The proteins in these cells, like in the embryos described above, also move around the cell depending upon events such as the stage of the cell cycle, whether or not the cells are polarized, state of differentiation, etc. To begin to understand the mechanisms by which these proteins are transported or sequestered will require additional analyses, including examination using high-resolution soft x-ray microscopy as described below. The cells will be fixed, the proteins labeled using our standard antibody labeling protocol, and the ultrastructural location of the proteins determined using cryo x-ray tomography. These analyses require the use of the cryo stage and tomography being developed in parallel with these experiments.

### *Cryo X-Ray Tomography*

A cryo-tilt stage has been developed in collaboration with the Center for X-Ray Optics. To test this apparatus, fixed *Drosophila* cells were placed in a micro-capillary tube approximately ten microns in diameter. The capillary tube enables collection of multiple projection images from multiple angles, with no obstruction. For the initial series, forty-eight projection images were collected at four-degree intervals. Tomographic reconstruction of these data was carried out to retrieve the three-dimensional data from these images, yielding structural information at approximately 50-nanometer resolution (see Figure 26). Further development is required to automate this process in order to increase the number of projections and to reduce the angle of separation, which will increase the resolution of the reconstructed data.



**Raw Data  
Projected image**



**Reconstructed Data  
Single section**



**Reconstructed Data  
Single section**

*Figure 26: Cryo X-Ray Tomography; Projected image and two sections from the reconstruction.*

## Material Sciences Division

---

### **Functional Bone-Like Materials: A Biomimetic Synthetic Approach**

---

Principal Investigators: Carolyn Bertozzi

Project No.: 01026

#### **Project Description**

The goal of the proposal is to develop new materials that are tough yet lightweight and fracture resistant. Such materials would find widespread use in industry. Nature creates a material—bone—that embodies these properties and our goal is to design artificial materials using natural bone as a guide. Bone consists of a microcrystalline inorganic composite called hydroxyapatite, embedded in an organic matrix that provides flexibility and elasticity. This paradigm will be recreated using synthetic constructs. Organic materials will be designed for micronucleation of inorganic crystals and the composites analyzed with respect to physical properties.

Organic microcrystalline materials will be generated from polymerized diacetylene arrays generated by self-assembly. Polydiacetylenes form various organized structures on the micron scale, such as tubes, liposomes, and ribbons, and these organized structures will be used to promote the crystallization of hydroxyapatite [ $\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2$ ], the ceramic found in natural bone. The microcrystalline nature of the organic scaffold will dictate the organization of inorganic crystals in the bulk material. The resulting composites will be further incorporated into a hydrogel polymer scaffold to provide physical shape and elasticity. Physical measurements of toughness and fracture resistance will be performed.

#### **Accomplishments**

Bone consists of microcrystalline hydroxyapatite and collagen, an elastic porous protein matrix that is decorated with mineral-nucleating phosphoproteins. Our rational design of artificial bone-like material uses natural bone as a guide. A hydrogel-based strategy is combined with a self-assembling polymer based strategy to assemble porous, functional three-dimensional organic scaffolds that are capable of template-driven mineralization of hydroxyapatite. Mineral infiltration techniques that are compatible with hydrogel formation are developed to effect the fabrication of the tailor-designed composite material.

Three libraries of monomers used for assembling functional organic scaffolds of the composite material were designed and synthesized. The first library of hydrogel monomers is methacrylamides that are functionalized with side chains carrying varied numbers and types of anionic groups. When polymerized, these anionic domains mimic the mineral nucleating ability of acidic matrix proteins in bone. The second library of hydrogel monomers carries either non-fouling hydrophilic side chain or adhesive peptide sequence that promotes specific cell-material interaction. Proper candidate(s) selected from these two libraries were combined and crosslinked (with various percentages of crosslinkers) via water-compatible radical polymerization to form hydrogels with novel physical property (e.g. elasticity, porosity), chemical versatility, and biocompatibility. Finally, to further control the order and conformation of the mineral nucleating ligand display on a nanoscopic level, a third library of self-assembling lipids was synthesized. They are bolaamphiphilic lipids functionalized with anionic surface residues. They self-assemble and polymerize into highly crystalline nano ribbons and sheets under ultraviolet irradiation. Under conditions compatible with hydrogel formation, these ordered polymeric mineral nucleating domains can then be covalently linked to the three-dimensional hydrogel network.

Several mineral infiltration techniques that are suitable for hydrogel mineralization have been developed. A homogeneous precipitation technique was applied to mineralize hydrogels and yielded unique microstructural patterns of hydroxyapatites that grew and tightly adhered onto the acidic hydrogels surface (Figure 27A). Alternatively, *in-situ* polymerization of the organic scaffold in the presence of hydroxyapatite yielded highly porous mineralized composites, with interconnected pore sizes ranging from 10s to 100s of microns (Figure 27B). Such porosity is in the same range of natural collagen membranes. Chemical and structural information of the composite materials was obtained at both micro- and nano-levels by Scanning Electron Microscopy and Energy Dispersive Spectrometry (SEM-EDS), High Resolution Transmission Electron Microscopy (HRTEM), and X-Ray Diffraction (XRD).

So far, we have demonstrated that functional organic-inorganic composite materials with necessary porosity and biochemical handles to promote cell adhesion and proliferation can be generated via a biomimetic synthetic approach. Mechanical properties of the composites are currently being evaluated. Optimization of mechanical property of these composites can be achieved by varying

chemical composition of the hydrogel, degree of crosslinking and mineral-to-gel ratio. Structural characterization of composites containing high order mineralization domains (formed by self-assembling

polymers) are also underway. Altogether, a new class of biomimetically designed artificial bone-like materials with improved properties over traditional bone implants are emerging.

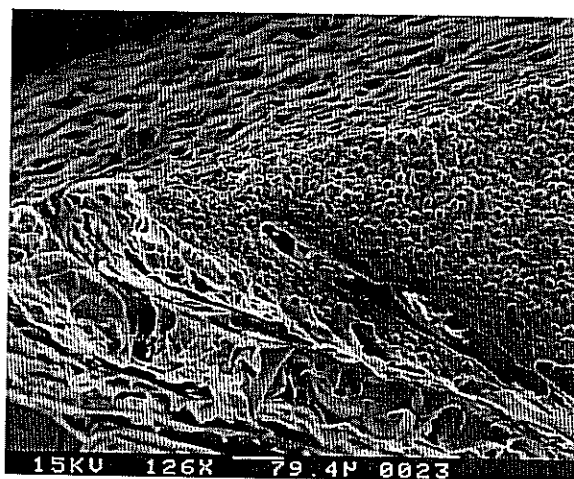
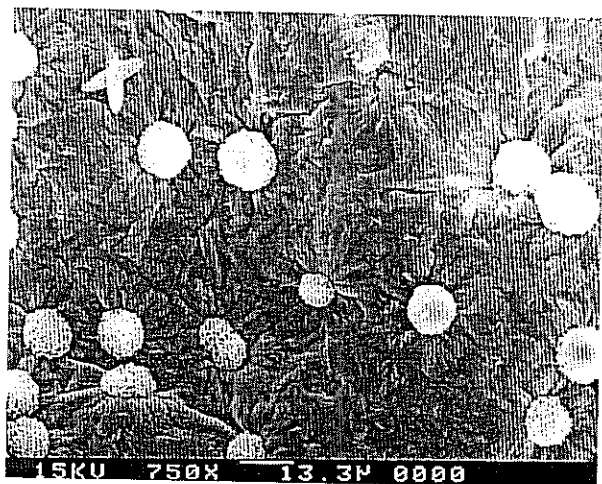


Figure 27: Scanning Electron Microscope (SEM) images of (A) hydroxyapatite grown on anionic hydrogel surface; (B) calcified porous anionic hydrogel.

## Publications

J. Song and C. Bertozzi, "A Biomimetic Approach toward Artificial Bone-like Materials," poster presentation at Gordon Research Conference: Biomaterials—Biocompatibility and Tissue Engineering (July 22-27, 2001).

J. Song and C. Bertozzi, "Functional Bone-Like Materials: A Biomimetic Synthetic Approach," abstract submitted to Spring 2002 MRS (Materials Research Society) Meeting, (April 1-5, 2001).

---

## Condensation of Indirect Excitons in Coupled Quantum Well Nanostructures

---

Principal Investigators: Daniel Chemla

Project No.: 01027

## Project Description

A system of spatially indirect (interwell) excitons in coupled quantum wells (CQWs) provides a unique opportunity for experimental study of two-dimensional (2-D) light bosons thermalized down to ultralow temperatures. The low-temperature, high-density excitonic matter is a potential candidate for observing Bose-Einstein condensation. Recent experimental studies of indirect excitons in CQWs have revealed nonclassical signatures seen in exciton transport and photoluminescence that are not explained within single exciton models and indicate possible coherency in the exciton system. In this project, we propose novel coherent spectroscopy experiments that



could reveal macroscopic quantum effects in the system of indirect excitons and, in particular, could identify the exciton condensation using new CQW samples specially designed for this project and novel experimental techniques with increased sensitivity to quantum coherence.

The coherence in an exciton system in CQW heterostructure can be directly detected by the coherence of the light it emits. We have prepared new AlAs/GaAs and GaAs/AlGaAs samples especially designed and optimized for this project. We propose to study the second and the fourth order correlation functions of the light emitted by these samples through: (1) (a) the speckles of Rayleigh scattering and (b) the study of the photon statistics by the measurement of the correlation of intensities registered by two different detectors; (2) the search for the excitonic insulator state by a time-resolved photoluminescence experiment; and (3) to realize experiments for exciton condensate in the traps, similar to the recent experiments for the atom Bose-Einstein condensate.

## Accomplishments

We are studying the system of indirect excitons in coupled quantum wells with emphasis on the problems indicated in the original proposal. In particular, the following progress has been achieved:

- The design work has been completed for the He3 refrigerator with optical access for spectroscopy measurements at temperatures down to 300 mK (including four-wave-mixing in transmitted and reflected geometry, resonant Rayleigh scattering, time-, angularly- and spatially-resolved photoluminescence). Currently the refrigerator is under production.
- New coupled quantum well sample for coherent optical spectroscopy of indirect excitons has been designed. The sample for the project has been grown by the group of Professor Gossard at University of California, (UC) Santa Barbara. Currently we process the sample by optical lithography in the Microfabrication Laboratory at UC Berkeley.
- We have performed the first measurements of the dispersion relation of a quasi-two-dimensional magnetoexciton. We demonstrated that the magnetoexciton effective mass is determined by the coupling between the center of mass motion and internal structure, and becomes overwhelmingly larger than the sum of the electron and hole masses in high magnetic fields. In particular, the observed mass enhancement explains the disappearance of the stimulated exciton scattering and the transition from the highly degenerate to classical exciton gas with

increasing magnetic field. The dispersion relations of indirect magnetoexcitons have been calculated. The experimental data are in a quantitative agreement with the *ab-initio* calculations.

## Publications

L.V. Butov, C.W. Lai, D.S. Chemla, Yu.E. Lozovik, K.L. Campman, and A.C. Gossard, "Observation of Magnetically Induced Effective Mass Enhancement of Quasi 2D-Excitons," *Physical Review Letters*, **87**, 21 (November 18, 2001)

<http://ojs.aip.org/getpdf/servlet/GetPDFServlet?filetype=pdf&id=PRLTA0000087000021216804000001>.

---

## Nanoscale Transport in Ultra-Thin Films

---

Principal Investigators: Michael Crommie

Project No.: 00018

## Project Description

As the size of electrical devices shrinks, the need to understand transport phenomena in microscopic structures grows. A basic understanding of material transport properties requires the characterization of electronic structure and defect configurations at the microscopic level. This is especially true of nanometer-scale devices, where electrical conductivity can be dominated by the action of only a single scatterer. The same electronic scattering processes that lead to voltage drops across a sample, however, also cause defect migration (electromigration) in current-carrying substrates. Electromigration is currently the largest single cause of failure in integrated circuitry, and will undoubtedly become even more of a problem as circuit size is reduced. A unified study of transport at the nanoscale should thus address both electrical conductivity and electromigration. Deeper understanding of electromigration has the added benefit of potentially transforming electromigration into a new tool for patterning surfaces and forming novel, non-equilibrium nanostructures. Such properties are of particular interest in new molecular film and nanocrystal samples.

We propose to fabricate a variable-temperature scanning tunneling microscope (STM) to study local transport phenomena in ultra-thin metal and molecular films. Future devices having nanometer-scale feature sizes are likely to be patterned from such films, and should thus reflect their transport properties. We intend to grow metal overlayers in

ultra-high vacuum (UHV) to give the cleanest, most well-characterized structures and surfaces. Electrical contact will be made to the films at four contact points so that accurate conductivity measurements can be made simultaneously with atomic-scale imaging in UHV. Scanning tunneling potentiometry will also be used to correlate local voltage drops with local film structure. Electromigration in the films will be studied by injecting current through the contact points and by monitoring changes in local structure with the STM. Optical access to the sample will also be possible in this system, allowing us to investigate the behavior of ultra-thin films and molecular assemblies in the presence of optical stimulation.

## Accomplishments

Over the last year we have completed the construction of a new variable temperature UHV STM with *in-situ* electron transport measurement capability and optical access. We tested the device under different conditions, and we are prepared to perform experiments on new molecular systems.

The STM utilizes a Pan-type coarse approach that is cooled within concentric heat shields using a gas-flow cryostat (temperature range is 15°K to 300°K). Our main progress was to integrate the STM into a UHV vacuum system with

sample preparation and analysis capabilities such as sputtering, annealing, Auger electron spectroscopy, and low energy electron diffraction (LEED) (see Figure 28). We also wired up the STM to homemade coarse approach electronics and commercial RHK scan electronics and software.

We have tested the STM on clean Au(111) surfaces over the temperature range 300°K to 40°K. The coarse approach works fine over this range, and we have observed clean terraces, step-edges, and the Au(111) herringbone reconstruction (see Figure 28). We have also tested the STM spectroscopy capability at room temperature and have succeeded in resolving the Au(111) Shockley surface state band-edge (see Figure 28). The STM has a low drift rate (less than six angstroms per hour at room temperature), but we are currently working to reduce vibrational and electronic noise arising from mechanical shorts and electronic groundloops.

With follow-on funding, we are now preparing to take data on a number of interesting physical systems, including optically active molecular adsorbates and novel silicon-on-insulator (SOI) heterostructures.

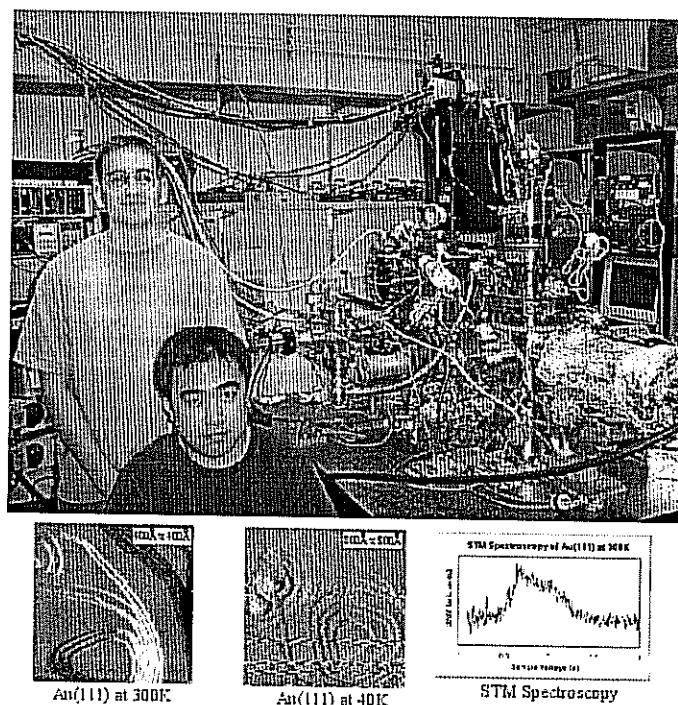


Figure 28: Katsumi Nagaoka and Matt Comstock stand near newly fabricated variable temperature UHV STM system. Preliminary data shown below includes images of Au(111) taken at 300°K and 40°K, as well as STM spectroscopy of Au(111) taken at 300°K.

---

## Atomically Resolved Spin-Polarized Imaging with Superconducting STM Tips

---

Principal Investigators: J.C. Séamus Davis

Project No.: 99024

### Project Description

The goal of this proposal is to create techniques of scanning tunneling microscopy (STM) with superconducting tips that will allow imaging of the spin polarization, and thus the magnetic moment, of individual atoms on a surface.

The project to make spin-polarized tips evolved into one to study the spin-polarized states at individual nickel impurity atoms. We have made a complete survey of this physics and have recently developed new applications of it in which we use the spin-polarized local states at nickel as nanoscale markers for superconductivity. These markers were applied in a study of nanoscale electronic phase separation and granular superconductivity in BSCCO, a highly correlated superconductor.

### Accomplishments

#### *Individual Magnetic Nickel Impurity Atoms*

In conventional superconductors, magnetic interactions and magnetic impurity atoms are destructive to superconductivity. By contrast, in some unconventional systems, e.g. superfluid  $^3\text{He}$  and superconducting  $\text{UGe}_2$ , superconductivity is actually mediated by magnetic interactions. A magnetic mechanism has also been proposed for high-temperature superconductivity (HTSC) in which an electron magnetically polarizes its environment resulting in an attractive pairing-interaction for oppositely polarized spins. Since a magnetic impurity atom would apparently not disrupt such a pairing-interaction, it has also been proposed that the weaker influences on HTSC of magnetic nickel impurity atoms compared to those of non-magnetic zinc are evidence for a magnetic mechanism. We used scanning tunneling microscopy (STM) to determine directly the influence of individual nickel atoms on the electronic structure of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ . Two local d-wave impurity-states are observed at each nickel. Analysis of their energies surprisingly reveals that the primary quasiparticle scattering effects of nickel atoms are due to *non-magnetic* interactions. Nonetheless, we also demonstrate that a magnetic moment coexists with unpaired superconductivity at each nickel site. The implications of

these phenomena, and those at zinc, are that the pairing-mechanism is magnetic in nature.

#### *Nanoscale Spatial Variations in Electronic Structure-YBCO*

Much experimental activity (and applications design) has focused on  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ , a material whose properties are, unfortunately, complicated by the presence of the  $\text{CuO}$  chain plane. We carried out measurements of the electronic structure of this plane from low-temperature scanning tunneling spectroscopy. The results show a gap in the electronic density of states (of magnitude  $\sim 25$  meV) that is filled by large numbers of intra-gap local-density-of-state (LDOS) resonances. This work is the first in which these resonances were discovered. Spatially, these resonances appear as incommensurate, one-dimensional LDOS oscillations along the  $\text{CuO}$  chains, while energetically they appear as intense peaks in the LDOS, which can occur with any energy within the energy gap. These electronic phenomena shed new light on recent results from other probes, and have strong implications for local electronic phenomena in the superconducting  $\text{CuO}_2$  plane, especially for nanoscale phase separation, e.g. stripes.

Understanding the electronic nature of the  $\text{CuO}$  plane, and its interaction with the  $\text{CuO}_2$  plane, may prove crucial to understanding the nature of superconductivity in  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$  (YBCO). At the very least, this plane, containing  $\text{CuO}$  chains running along the  $b$ -axis, breaks the tetragonal symmetry of the YBCO crystal by introducing a one-dimensional structure. This complicates the search for one-dimensional spatial ordering of magnetic and electronic structure (striping) in the  $\text{CuO}_2$  plane that has been predicted to exist in the cuprate high-temperature superconductors. These one-dimensional structures, "stripes", are believed to arise from the spatial segregation of two competing electronic phases, antiferromagnetism (AF) and superconductivity (SC), and are usually described as insulating AF regions separated by hole-rich SC domain wall. We have shown there are other strong one-dimensional physical phenomena that are not "stripes".

#### *Spatial Inhomogeneity in LDOS of Optimal BSCCO*

In collaboration with angle resolved photoemission spectroscopy (ARPES) researchers at Boston University and Boston College, we show that the spatial inhomogeneity in the gap magnitude is correlated with the integrated density of electronic states. That means that the regions with high hole-density are possibly better superconductors than those that are not. We speculate that this may be a fundamental issue in HTSC whereby the dopant atoms create disorder at the same time as they introduce holes into the  $\text{CuO}_2$  plane.

### *Evidence for Granular Superconductivity in Underdoped in BSCCO*

High-temperature superconductivity appears in the cuprate-oxides when holes are introduced into the  $\text{CuO}_2$  crystal planes. For low hole-densities, theory has long predicted that nanoscale electronic phase separation (EPS) should occur. Recent research has focused on one-dimensional "stripes" as exemplifying EPS, but it could also be manifest as hole-rich superconducting "domains" surrounded by another electronic phase. Although magnetic phenomena exist at low hole-densities which may be consistent with nanoscale EPS, their direct electronic signatures have proven elusive. To search for such signatures in underdoped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ , we performed comprehensive high-resolution studies of the spatial interrelationships between tunneling spectra measured by STM. They reveal an apparent spatial segregation of the electronic structure into  $\sim 3$ -nanometer diameter domains (with superconducting characteristics and energy gap  $\Delta$  always less than 50 meV) embedded in an electronically distinct background. To explore whether this situation represents EPS, we next employed scattering resonances at nickel impurity atoms as nanoscale "markers" for the local existence of superconductivity. No nickel-resonances are detected in any regions with local energy gap  $\Delta > 50 \pm 2.5$  meV. The disappearance of both the apparently superconducting domains and the nickel quasiparticle scattering resonances wherever  $\Delta > 50$  meV implies a local electronic structure in these regions that is significantly different. This is a breakthrough because we have in fact observed granular superconductivity in an HTSC crystal for the first time by using the nickel impurity states as "markers".

### **Publications**

S.H. Pan, E.W. Hudson, A.K. Gupta, K-W Ng, H. Eisaki, S. Uchida, and J.C. Davis, "STM Studies of the Electronic Structure of Vortex Cores in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ ," *Physical Review Letters* **85**, 1536 (2000).

E.W. Hudson, K.M. Lang, V. Madhavan, S.H. Pan, H. Eisaki, S. Uchida, and J.C. Davis, "Interplay of Magnetism and High- $T_c$  Superconductivity at Individual Magnetic Impurity Atoms in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ ," *Nature*, **411**, 920 (2001).

S.H. Pan, J. O'Neil, R.L. Badzey, C. Chamon, H. Ding, Z. Wang, H. Esiaki, S. Uchida, E.W. Hudson, K.M. Lang, and J.C. Davis, "Microscopic Electronic Inhomogeneity in the High-Temperature Superconductor  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ ," *Nature*, **413**, 282 (2001).

---

### **Surfactant-Mediated Epitaxy of IV-IV Compounds: Expanding the Limits of Alloy Composition and Nanostructure Synthesis with Lead Overlayers**

---

Principal Investigators: Oscar Dubón, Jr.

Project No.: 01028

### **Project Description**

The presence of a surface layer of segregating impurities during epitaxial growth can produce changes in the morphology of the film. Commonly called "surfactant," the impurity overlayer changes the arrangement of substrate adatoms and the mobility of depositing atoms and therefore represents an experimental parameter that can be exploited to influence growth kinetics. Control over growth kinetics is an essential ingredient for the effective manipulation of self-organizing processes that lead to periodic semiconductor nanostructures including island arrays and alloy superlattices.

A layer of lead atoms will be used in the molecular beam epitaxial growth of group-IV-based alloys in order to probe surfactant effects on spontaneous pattern formation. The choice of lead as a surfactant is based on recent experiments showing that a lead layer in germanium and silicon thin-film epitaxy dramatically reduces the minimum temperature for epitaxial growth. Unlike other commonly used surfactant species, lead does not degrade the electrical properties of the films. The lead layer can mitigate surface segregation of the alloy constituents enabling the exploration of alloy compositions that up to now have not been accessible. Alloys highly enriched with the element tin are the primary focus of our studies. These materials provide novel possibilities for the study of alloy compositional modulation in highly mismatched systems. Expanding the limits of alloy composition widens the range of lattice-mismatch-induced strain fields that can be established. In addition, passivation of the sample surface by the lead layer can result in pronounced changes in the surface mobilities of the depositing species. These effects will have a significant impact on the spontaneous formation of quantum dot arrays and may provide an important avenue to influence the self-organization process in a controlled manner.

## Accomplishments

The first phase of the proposed work consisted of establishing advanced ultra-high vacuum deposition capabilities. To this end we have rehabilitated a RIBER 32 molecular beam epitaxy system. Ultra-high vacuum in the epitaxy chamber has been achieved with a sustained base pressure at or below  $1 \times 10^{-10}$  torr. We have developed significant capabilities for the deposition of silicon, germanium, tin, and lead, both sequentially and simultaneously. Epitaxy can be monitored *in situ* using a state-of-the-art reflection high-energy electron diffraction (RHEED) system capable of recording the morphological evolution of the deposited layers.

We have deposited a variety of thin-films. Thus far, we have grown silicon and germanium homoepitaxial films of high structural quality. In addition, we have initiated investigations on the growth of SiGeSn alloys on germanium substrates. We have found that about 1 atomic percent of tin can be incorporated at substitutional lattice sites when the substrate is held at a temperature of 300 °C. These alloys are of great interest because the entire range of strain conditions for a deposited film can be achieved depending on the composition of the film, from tensile to fully relaxed yet coherent to compressive. The successful epitaxial growth of these materials is an essential step toward the investigation of surfactant effects on self-organizing processes during growth.

---

## Holographic Imaging with X-Rays: Fluorescence and Fourier Transform Holography

---

Principal Investigators: Charles Fadley and Malcolm Howells

Project No.: 00030

## Project Description

The purpose of this project is to develop two promising new techniques for imaging matter in three dimensions using x-rays. The first technique is hard x-ray fluorescence holography, which has already demonstrated the ability to image atomic positions in three dimensions with accuracies of  $\sim 0.2$  angstrom over a range of about 10 angstroms around a given emitter. This will be extended to 0.1-angstrom resolution over larger spatial regions and to yield

enhanced elemental and magnetic sensitivity. The second technique is soft x-ray Fourier transform holography, which currently is able to image wet life-science samples at 50-nanometer resolution in two dimensions. This will be further developed to yield ultimate resolutions of  $\sim 10$  nanometers in three dimensions.

To accomplish this, an existing x-ray fluorescence holography experimental station will be upgraded in several ways for use at the Advanced Light Source (ALS), first, on a bend-magnet beamline (9.3.1), and, ultimately, on a superbend or wiggler beamline in the future. The x-ray energy discrimination and detection will be via high-speed solid-state detectors (making use of special Berkeley Lab expertise). Experiments will be performed in both the direct (single-energy) and inverse (multi-energy) modes, as well as with resonant absorption, to enhance contrast, elemental sensitivity, and magnetic sensitivity. An existing soft x-ray holography experimental station will also be modified to permit the use of ALS beamline 9.0.1 for Fourier transform holography with a complex reference object (e.g., a random array of ultrasmall pinholes). New theoretical methods for inverting both hard x-ray and soft x-ray Fourier transform holograms will also be explored, and there will be a high degree of synergism between the two aspects of this work.

## Accomplishments

### X-Ray Fluorescence Holography (XFH)

A new experimental system for XFH at the Advanced Light Source has been developed, optimized for the energy range of the relevant beamline, and used successfully to measure first holograms and holographic atomic images. The experiments were performed on beamline 9.3.1, which was originally designed to work at relatively low energies ( $<5$  keV), but was reconfigured for our work to operate at up to  $\sim 7$  keV. The experimental chamber, under low vacuum ( $\sim 10^{-4}$  Torr), is separated from the high vacuum of the beamline ( $10^{-9}$  Torr) by a beryllium window. Inside the chamber, the sample is mounted on a two-axis goniometer which permits very rapid angle scanning in both the azimuthal (3600 degrees per second), and polar (2 degrees per second) directions, thus permitting the acquisition of a full hologram over a nearly  $2\pi$  solid angle in about 40 seconds. Data acquisition and system diagnostics are controlled by dedicated software and electronics developed by us. The measurement is repeated over a few hours until the statistical noise and effects of incident beam fluctuations have been reduced to acceptable levels.

With this system it has proven possible to reliably measure holograms with modulations of only a few tenths of a percent for several materials to date. These materials include simple structures such as MnO in (001) surface orientation (Figures 29a-c) and vanadium in (111) orientation, and more complex structures that will be

further studied in the future—a colossal magnetoresistive (CMR) material  $[(\text{La}_{1-x}\text{Sr}_x)_3\text{Mn}_2\text{O}_7]$  (Figure 29d) and a fivefold-symmetric quasicrystal (AlPdMn). The only limitation in the system, for some samples with larger unit cells, is that illumination of the solid state detector by a Bragg reflection from the sample can cause the detector to become saturated, making the hologram more difficult to interpret. A new detector system using a graphite analyzer is being planned to surmount this problem.

Beyond this experimental work, we have developed several theoretical methods for extracting further structural information from experimental holograms: (1) Resonant-XFH (RXFH), which is based on the anomalous scattering effect currently used in normal x-ray diffraction to solve the phase problem. Using this new technique, we can suppress non-resonant atoms and image only selected atomic species, yielding images “in true color”; (2) Holographic determination of structure factor phases, in which analyzing the holograms in reciprocal space has yielded the phases of the structure factors of a vanadium crystal; (3) New imaging algorithms based on iterative deconvolution methods, which have demonstrated resolutions better than the diffraction limit; and (4) Differential photoelectron holography (DPH), in which two holograms at slightly different wavelengths are subtracted so as to suppress forward scattering effects, thus greatly improving image quality. This method also will be applied to x-ray fluorescence holography in the future.

### Soft X-Ray Holography (SXH)

The development of new types of reference objects and the use of the Fourier transform geometry will enable high resolutions to be obtained with a relatively low-resolution detector via diffraction tomography. Such a reference

object must have good x-ray transmission and possess diffraction structure on the interesting  $0.1$  to  $0.001 \text{ \AA}^{-1}$  scale, such that it fills the detector with diffracted x-rays out to the desired angle. During this grant period, we first made experimental tests of pinhole arrays and silica aerogels as candidate reference objects. These test objects were illuminated by unmonochromatized undulator radiation (“pink beam”) on beamline 9.0.1 at the Advanced Light Source. During the first part of this year, we established a new experimental apparatus, which is compatible with the hardware of the beam line and its other users. We have also completed an upgrade program for the branch line, which allows (but does not require) the introduction of a new component (a Fresnel zone plate) to provide improved monochromatization of the beam to about  $0.1\%$ . Although the new monochromator was working at the end of our last run, two of our five zone plates have been destroyed, apparently by the intense beam. Thus, some further work to improve the durability of the zone plate is required and will be undertaken before the next run. We have now had two running periods, three shifts in July and ten in November. The first was spent commissioning the experimental apparatus and the second was used to commission the monochromator and to establish the technique of recording very high resolution soft x-ray diffraction patterns on our charge coupled device (CCD) detector. The latter was successfully achieved using a test object made of 20-nanometer gold balls for which the diffraction pattern was recorded out to an angle corresponding to a resolution better than  $100 \text{ angstroms} = 10 \text{ nanometers}$  (see Figure 29e). Subsequent experiments in the latter cycle and the May-to-December 2002 cycle will focus on use of improved zone plates, better coherence control, and more interesting test samples for which images can be reconstructed by phase retrieval.

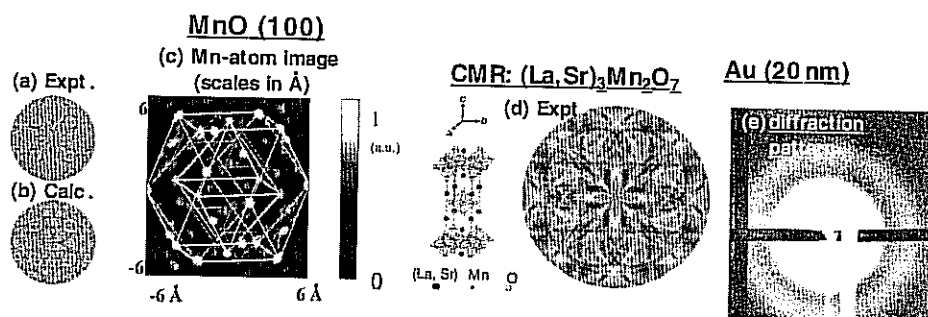


Figure 29: (a) Experimental hologram from an MnO (001) sample at 6.93 keV: a total of  $10^{11}$  photons were collected over  $3.2 \times 10^5$  data points. (b) Simulated MnO hologram. (c) Experimental Mn-atom reconstruction: ~50 atoms are visible. (More details and animated image at <http://electron.lbl.gov/marchesini/als/indexnew.html>). (d) Crystal structure and hologram from a CMR sample. (e) Diffraction pattern of 560 eV x-rays scattered from 20-nanometer gold balls, as recorded on a CCD.

## Publications

S. Opmori, L. Zhao, S. Marchesini, M.A. Van Hove, and C.S. Fadley, "Resonant X-Ray Fluorescence Holography: Three-Dimensional Atomic Imaging in True Color," *Physical Review B*, **65**, 014106 (2002).

S. Marchesini, N. Mannella, L. Zhao, *et al.*, "Holographic Determination of Structure Factors," draft in preparation for *Physical Review B*.

S. Marchesini, C.S. Fadley, *et al.*, "X-Ray Fluorescence Holography: Beyond the Diffraction Limit," draft in preparation for *Physical Review B*.

S. Opmori, Y. Nihei, E. Rotenberg, J.D. Denlinger, *et al.*, "Differential Photoelectron Holography: A New Approach for Three-Dimensional Atomic Imaging," *Physical Review Letters* (in press).

M.R. Howells, C.J. Jacobsen, S. Marchesini, S. Miller, J.C.H. Spence, and U. Weierstall, "Toward a Practical X-Ray Fourier Holography at High Resolution," *Nuclear Instruments and Methods A*, **468**, 864-867, (2001).

J.C.H. Spence, M.R. Howells, L.D. Marks, and J. Miao, "Lensless Imaging: A Workshop on 'New Approaches to the Phase Problem for Non Periodic Objects'," *Ultramicroscopy* (accepted for publication).

U. Weierstall, Q. Chen, J.C.H. Spence, M.R. Howells, and R.R. Panepucci, "Image Reconstruction from Electron and X-Ray Diffraction Patterns using Iterative Algorithms: Theory and Experiment," *Ultramicroscopy* (accepted for publication).

---

## A Molecular Foundry: Complex Nanosystems for Designed Structure and Function

---

Principal Investigators: Paul Alivisatos and Jean Fréchet

Project No.: 01029

## Project Description

Composites of inorganic nanocrystals and organic polymers are particularly interesting materials in the study of electrical transport. The band gaps and offsets of typical semiconducting nanocrystals and polymers are such that charges separate across an interface between them. The purpose of this project is to create effective

solar cells comprised a layer of cadmium selenium (Cd/Se) nanocrystals/poly(3-hexyl thiophene) composite between two electrodes of Indium Tin Oxide (ITO) and aluminum.

Upon sun exposure, excitons are formed both in the semiconductor nanocrystal and polymer matrix. Such excitons migrate in their respective media and may lead to charge-separation if they encounter an interface nanocrystal/polymer. For instance, if the exciton is created on the nanocrystal, the hole can subsequently transfer to the polymer, producing a charge-separated state with an electron on the nanocrystal and a hole on the polymer.

Important factors affecting the performance of the photovoltaic device include the overlap of the device absorption with the solar spectrum, and the efficiency of carrier separation and transport. The broad absorption of the Cd/Se nanocrystals can be tuned by changing their size/shape, and efficient charge separation may be obtained by incorporating semiconductor surfactants at the nanocrystal/polymer interface. Such Cd/Se nanocrystal/semiconductive polymer composites have the advantage of being easy to process (spin-casting films) to form large-area devices.

## Accomplishments

Without surfactants, Cd/Se nanocrystals are insoluble in organic matrices. To the best of our knowledge, all surfactants used in combination with Cd/Se nanocrystal consist of alkyl phosphine oxides, e.g. trioctyl phosphine oxide or alkyl amine. Such surfactants are inappropriate for solar cell devices since they form a thin insulator layer around the nanocrystal quenching any physical electron transfer at the nanocrystal/polymer interface. Therefore, we designed and synthesized a series of electroactive surfactants to allow transfer and accumulation of electrons into the Cd/Se nanocrystals, and holes in the polymer matrix.

Such surfactants consist of essentially two units: a P-based moiety with high affinity toward the Cd/Se nanocrystal, and a semiconducting thiophene unit facing the polymer matrix. These molecules have been produced using syntheses of 3 to 12 steps depending on the surfactant complexity. Each new semiconducting compound has been fully characterized by <sup>1</sup>H-Nuclear Magnetic Resonance (NMR), <sup>13</sup>C-NMR and mass spectroscopy.

In presence of such surfactants, the initially insoluble Cd/Se nanocrystals are found to be soluble and they can be dispersed well in poly(3-hexyl thiophene).

Moreover, photoluminescence spectroscopy study on these surfactants-coated Cd/Se nanocrystals has successfully shown that electron transfer occurs from the

surfactant to the nanocrystal via a Dexter mechanism (physical electron transfer).

This encouraging result led us to fabricate a full device containing all the required elements. This device is currently undergoing photophysical characterization.

Also, our ongoing investigation should lead to a new generation of electroactive surfactants that provide optimized nanocrystal surface coverage for maximum efficiency.

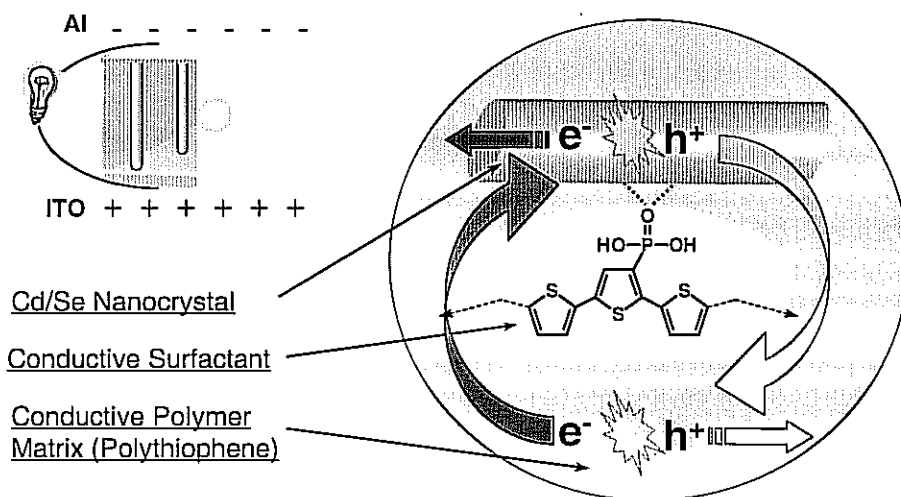


Figure 30: Schematic representation of the solar cell device based on Cd/Se nanocrystal/polythiophene composite: under sun irradiation, excitons are formed in the nanocrystal and in the polymer. The specially designed electroactive surfactant enables charge separation to occur by electron transfer from the polymer to the nanocrystal leading to an overall electric field at the ITO/Al electrodes.

## Publications

T. Emrick, C. Pitois, and J.M.J. Fréchet, "Functional Quantum Dots: Growth of Cadmium Selenide Nanocrystals in Di-n-octylundecenyl Phosphine Oxide (DOUPO)," (draft).

C. Pitois, D. Milliron, W. Huynh, J.M.J. Fréchet, and P. Alivisatos, "Role of Electroactive Surfactants in Cd/Se Nanocrystals/Poly(3-hexyl thiophene) Composite Photovoltaic Device," (draft).

## High Pressure Photoelectron Spectroscopy for Catalysis, Semiconductor Processing, and Environmental- and Bio-Chemistry

Principal Investigators: Miquel Salmeron, Frank Ogletree, Charles Fadley, and Zahid Hussain

Project No.: 99028

## Project Description

A High Pressure Environmental Photoelectron Spectrometer (HPEPS) has been developed over the last three years at the Advanced Light Source (ALS). The HPEPS is a unique instrument, the only one of its kind in the world, making the ALS the only place where photoelectron spectroscopy can be performed on surfaces at gas pressures above 1 Torr. The lens system was designed and fabricated in FY 1999. Assembly, testing, and initial measurements, including determination of



operating parameters, were carried out in FY 2000. Prior year annual reports give further details.

We demonstrated the technical feasibility of the HPEPS system in FY 2000. Our goal for FY 2001 was to demonstrate the scientific feasibility of the HPEPS system by constructing a sample chamber to carry out a series of demonstration experiments under controlled conditions, and obtain new scientific results in several important areas of contemporary surface science.

## Accomplishments

All of the major goals for FY 2001 have been met:

- The experimental cell of the HPEPS system was improved to allow for better control of reactant gases and sample temperature. The cell design was modified to be truly ultra-high vacuum compatible and the x-ray window changed from silicon nitride to aluminum. After bakeout the cell base pressure is now in the 10 to 10 Torr range, which now allows experiments to be carried out with well-defined sample surface conditions.
- A series of experiments to study the premelting of the ice surface, initiated in FY 2000, were completed. The hydrogen bonding of water molecules was probed using near-edge x-ray adsorption fine structure (NEXAFS) measurements of ice and water surfaces in equilibrium with water vapor and the temperature dependence of premelting was investigated. Surface sensitivity was obtained using Auger electron detection. Initial measurements in FY 2000 indicated that surface hydrocarbon contamination influenced premelting. With the improved sample, cell data for chemically clean ice surfaces were obtained and the role of contamination was clarified.
- A series of measurements to study the catalytic oxidation of methanol to formaldehyde over copper oxide under reaction conditions was carried out in collaboration with the group of Professor R. Schloegl of the Fritz Haber Institute of the Max Planck Gesellschaft in Berlin. Previous studies suggested that a "copper sub-oxide" played a key role in the reaction. The partial pressures of the oxygen and methanol reactants were varied in the Torr range as the sample temperature was varied. The sample electronic structure was monitored using *in-situ*, high-resolution x-ray photoemission spectroscopy (XPS) while the reaction rate was

monitored by mass spectroscopic analysis of the gases escaping from the reaction cell.

- Experiments on the co-adsorption and oxidation of CO and NO over Rh(111) have been initiated. This is an important catalytic system for environmental applications (automotive exhaust catalysis).
- The success of the HPEPS concept has led to a modification of the existing Molecular Environmental Science (MES) Beamline project. This project originally included two endstations, a scanning x-ray microscope and a photoemission spectroscopy chamber. The MES project has been expanded to include a dedicated HPEPS endstation. This will be a "second generation" high pressure spectrometer based on the results obtained through this Laboratory Directed Research and Development (LDRD) project.

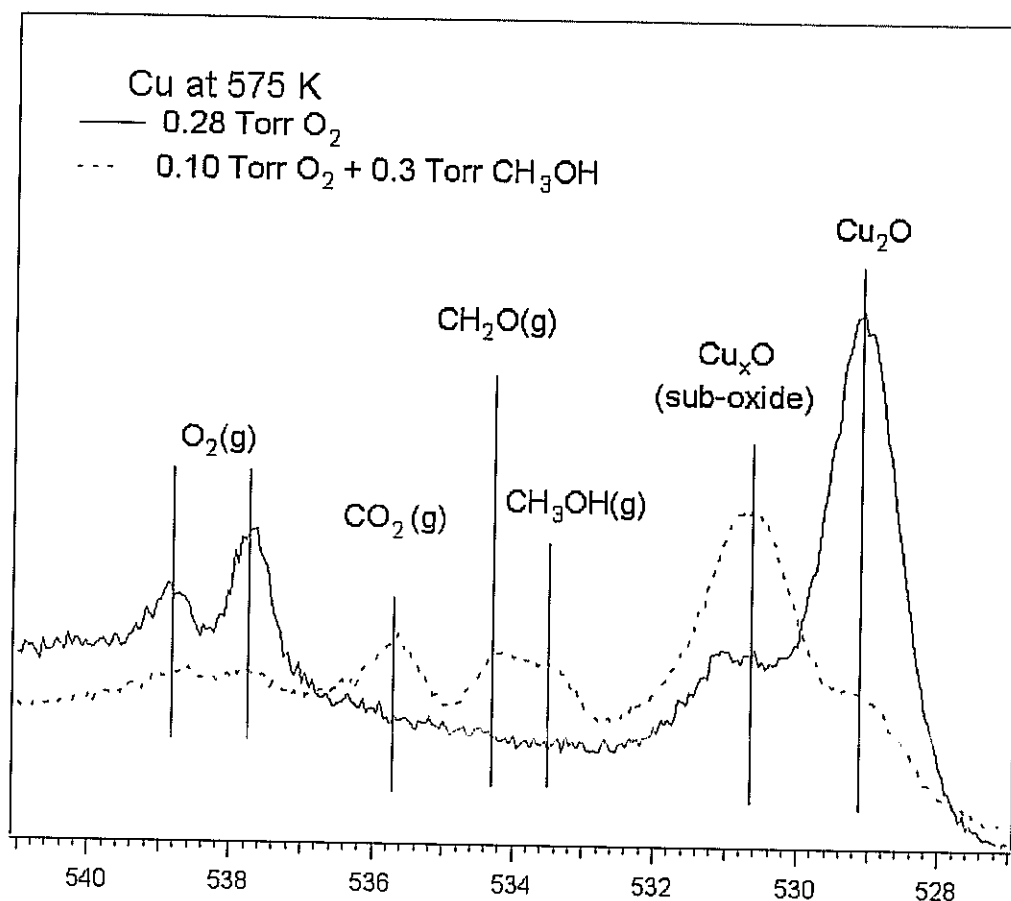


Figure 31: X-ray photoemission spectrum of the oxygen 1s-core level photoemission region of a copper oxide catalyst during methanol oxidation. Both surface oxygen species as well as gas phase reactants (g) and products can be observed.

## Publications

H. Bluhm, M. Havecker, A. Knop-Gericke, D.F. Ogletree, M. Salmeron, V.I. Bukhtiyarov and R. Schlögl, "In Situ XPS Investigation of Methanol Oxidation Over Copper," (in preparation).

H. Bluhm, D.F. Ogletree, F. Requejo, C.S. Fadley, Z. Hussain, and M. Salmeron, "The Premelting of Ice Studied with Photoelectron Spectroscopy," to be submitted to *Journal of Chemical Physics*.

H. Bluhm, M. Havecker, A. Knop-Gericke, F. Requejo, D.F. Ogletree, C.S. Fadley, Z. Hussain, M. Salmeron, and R. Schlögl, "High Pressure XPS: A New Tool for the In-Situ Investigation of Catalysts," delivered to American Vacuum Society International Symposium, San Francisco, (October 2001).

D.F. Ogletree, H. Bluhm, G. Lebedev, C. Fadley, Z. Hussain, and M. Salmeron, "A Differentially Pumped Electrostatic Lens System for Photoelectron Spectroscopy Studies in the Torr Pressure Range," draft, to be submitted to *Applied Physics Letters*.

---

## **Laser Spectroscopy on New Materials: Theory and Experiment**

---

Principal Investigators: Yuen-Ron Shen

Project No.: 01030

### **Project Description**

Semiconducting and magnetic nanostructures, quantum dots, composite materials, organic and polymeric films, chiral molecules and systems, and magneto-optoelectronic materials have attracted increasing attention. While optics is one of the most effective means for material studies, few laser spectroscopic techniques have been developed and applied to such materials. The goal of this project is to understand nonlinear optical processes in these materials and then develop corresponding techniques to study these materials. Being far more versatile than the linear ones, the nonlinear optical probes are expected to yield a wealth of information about both the equilibrium and the dynamic properties of the materials. Work involves theoretical and experimental research in collaboration with Professor Christos Flytzanis and his group at Ecole Normale Supérieure in Paris, France.

### **Accomplishments**

Nonlinear optical spectroscopic study of chiral materials has been initiated. Optical sum-frequency generation process in chiral liquids, predicted 35 years ago, was observed for the first time. It exhibits resonance enhancement as the output sum frequency approaches electronic resonance. This shows that the process can be used as a novel method to probe molecular chirality of a chiral medium for chemical and biological applications.

In collaboration with Professor Christos Flytzanis, a microscopic model calculation was carried out to relate the chiral response deduced from sum-frequency generation to the stereochemical features of the molecules.

A research program on studies of materials using nonlinear polarization-sensitive optical processes was initiated. A sensitive up-conversion scheme for detection of time-resolved polarized infrared luminescence was developed. Investigation of spin dynamics in semiconductors (with Peter Yu's group) and magnetic materials (with Z. Qiu's group) is being planned.

Photo-induced effects in media were analyzed. It was recognized that a circularly polarized photo-excitation could induce both a magnetization and a change of

chirality in a medium. A model calculation is being carried out to illustrate the effect more quantitatively.

### **Publications**

M. Belkin, S. Han, and Y.R. Shen, "Sum-Frequency Generation in Chiral Liquids: Electronic Case," *Physical Review Letters* **87**, 113001 (September 2001).

M. Belkin, Y.R. Shen, and C. Flytzanis, "Sum-Frequency Spectroscopy on Binaphthyl Solution near Excitonic Resonance: A Model Calculation," (in preparation).

---

## **Single-Molecule Protein Dynamics**

---

Principal Investigators: Shimon Weiss

Project No.: 99030

### **Project Description**

This project seeks to develop methods to study conformational changes and folding/unfolding pathways of single macromolecules by novel single-molecule fluorescence spectroscopy techniques. Fluorescence resonance energy transfer (FRET), which measures the proximity between two fluorophores, is used to measure distance changes, and therefore conformational dynamics, between two points on a macromolecule. This distance serves as a reaction coordinate for folding/unfolding, denaturation and charge screening reactions. Performing such measurements on the single molecule level has important advantages over conventional ensemble measurements, since it (1) resolves and quantitatively compare distinct sub-populations of conformational states, otherwise invisible at the ensemble level; and (2) resolves dynamic conformational changes, otherwise hidden at the ensemble level because of lack of synchronization.

Two kinds of molecules have been investigated: single-stranded DNA (ss-DNA) and the protein chymotrypsin inhibitor 2 (CI2). In the first experiment, ss-DNA is used to study the statistical and dynamical properties of a short polymer, with only very few Kuhn segments. While the conformational statistics of long single- and double-stranded DNA have been studied extensively using traditional methods such as sedimentation velocity, light scattering, and optical tweezers, these methods are inadequate for the studies of polymers on the 100 angstrom scale, far from the infinite chain limit. Properties such as the persistent length and the characteristic ratio of

DNA oligonucleotides in this distance range (15 to 60 bases in length) change significantly with increasing chain length. While models have been proposed to calculate the end-to-end distance distributions and scaling properties in such oligonucleotides, they have not been systematically tested experimentally. Such scaling issues and the effects of solvent environment on the polymer conformational properties are being studied.

The molecule CI2 is used for single-molecule protein-folding experiments. The problem of protein folding is of great significance from both fundamental and practical points of view. How a relatively unstructured protein in its unfolded state finds its way through a huge conformational space to a small ensemble of structures that is the native state is a question that has fascinated physicists, chemists, and biologists for many decades. Furthermore, the current effort in genome research is producing massive quantities of protein sequence information that needs to be converted into structural and functional information. A detailed understanding of protein-folding mechanisms is therefore a very important piece of this puzzle. Although much has been learned about the issue, especially in the last decade, the complexity of protein structure precludes detailed studies from being carried out using ensemble methods. These methods lose a lot of the dynamic and stochastic information that is both fascinating and crucial to the understanding of the physics of protein-folding mechanisms.

## Accomplishments

A universal feature of all protein-folding reactions is the collapse of the polypeptide chain from its expanded denatured structure to a more compact native-like structure. A fundamental question arises as to how the above collapse and specific structure formation occur. What are the general structural features, either in specific sequence, or more generally in topology that governs the kinetics of these two events? Do the two occur sequentially, or are they coupled to each other?

Towards this end, we have extended single-pair fluorescence resonance energy transfer (spFRET) measurements to include fluorescence-lifetime information obtained by time-correlated single photon counting (TCSPC) and applied this enhanced methodology to study the folding reaction of chymotrypsin inhibitor 2 (CI2).

For TCSPC, a pulsed laser is used to excite a sample, and the time between a photon's arrival and the next laser pulse is measured with sub-nanosecond accuracy. This is called the "microscopic" arrival time. Fluorescence-lifetime decay histograms formed from binning the photons by their microscopic arrival times are used to obtain information about the fluorescence lifetime of the

fluorophore under investigation. The arrival time of the photon is also measured with respect to a global clock (100-nanosecond accuracy). This is called the "macroscopic" arrival time. The relative timing of the photons is then known, allowing photon bursts from single molecules to be sifted from the data (this is the first step in the solution of single-molecule data analysis.) The raw data is thus a list of photons, each tagged with the detector channel, the microscopic arrival time, and the macroscopic arrival time. After sifting photon bursts from the data, the next task is to calculate the various quantities of interest from these bursts. Simple statistics include the number of photons in each detector channel and the width of the burst. As described previously, energy transfer efficiency can be calculated from the intensity of the donor and acceptor channels, the background levels, and the misalignment factor  $\gamma$  (see last year's Laboratory Directed Research and Development report). In addition, the lifetime of each channel can be calculated from the data. In particular, the donor lifetime can be calculated, obtaining an independent estimate of energy transfer efficiency ( $E$ ).

In addition to the ability to measure  $E$  without calibrating the factor  $\gamma$ , there is an additional benefit to using lifetime information with spFRET: when the donor lifetime is used in conjunction with  $E$  calculated from the intensity ratio, there is an improvement in the ability to separate subpopulations. This can be seen in the attached figure. The one-dimensional histogram of  $E$  from intensity alone does not separate the subpopulations as well as the two-dimensional histogram shown.

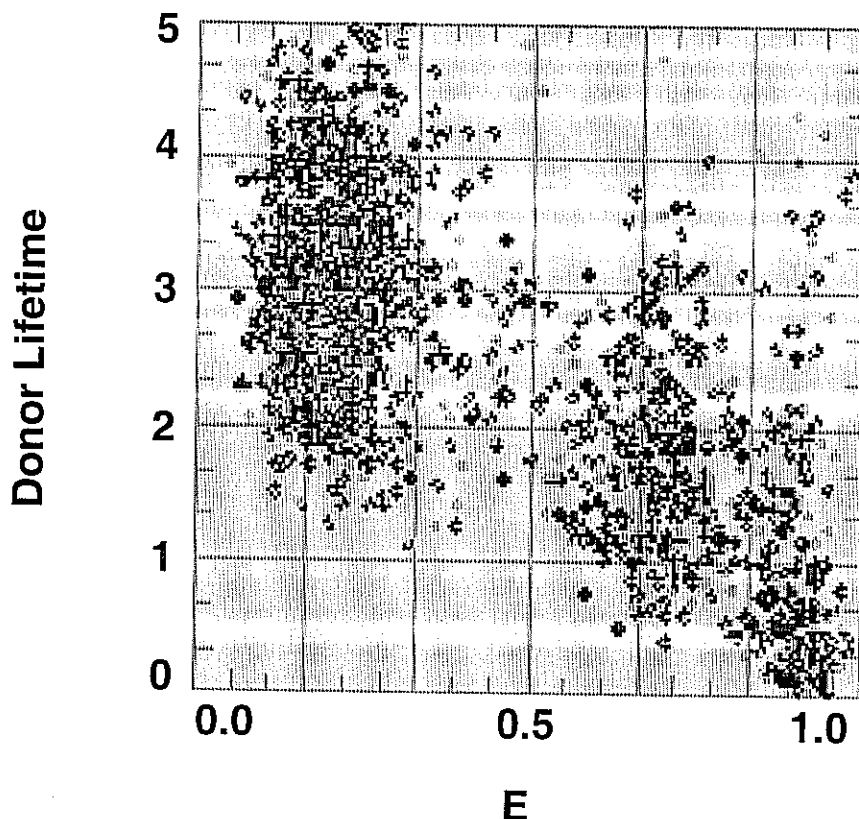


Figure 32: The two-dimensional histogram shows the number of bursts with a given donor lifetime and FRET efficiency  $E$  (calculated using intensity.) The sample is the doubly labeled C12 molecule discussed previously, in a denaturant concentration of 4 molar guanidine hydrochloride (GuHCl). The folded population is visible at  $E > 0.8$ . The unfolded population is visible between  $0.5E$  and  $0.8E$ . The "zero peak," with inactive or photobleached Cy5 is also visible in the upper left corner. This population is used to obtain the donor lifetime in the absence of the acceptor.

### Publications

A.A. Deniz, T.A. Laurence, M. Dahan, D.S. Chemla, P.G. Schultz, and S. Weiss, "Ratiometric Single-Molecule Studies of Freely Diffusing Biomolecules," *Annual Reviews in Physical Chemistry*, **52**, 233-253 (2001).

### *Infinite $[Mo_3Se_3]^-$ Chains as Molecular Conductors and Their Assemblies*

Principal Investigators: Peidong Yang

Project No.: 01031

### Project Description

The rational organization of molecular wires into two-dimensional or three-dimensional assemblies with novel functional properties is of critical importance to the development of emerging molecular electronics. A fundamental question is what type of interactions between molecular wires are essential for their organization into

ordered two-dimensional and three-dimensional assemblies and how can we rationally tune these interactions, and eventually their electronic properties. This research project is aimed at (1) developing general strategies for organizing molecular wires into two-dimensional and three-dimensional assemblies and circuitries; (2) understanding the underlying organization mechanism under either equilibrium or non-equilibrium conditions; and (3) exploring the electron transport behavior for the individual wires and their assemblies. This study will serve as rational basis for designing and creating new nanostructured materials based on molecular wires and exploring systematically their potentially unique properties.

$[\text{Mo}_3\text{Se}_3]_\infty$  infinite molecular chains are chosen as our experimental model system because of their high conductivity and chemical accessibility. Mesoscopic organization of metallic  $[\text{Mo}_3\text{Se}_3]_\infty$  chain in the presence of cationic surfactants will be explored first. Structural ordering including formation of lamellar and hexagonal mesophases is expected to evolve from the complex long-range repulsive and/or short-range attractive interactions within the systems. We will also explore the possibility of organizing these wires into novel electronic configurations and architectures on substrates. Two approaches will be explored. One relies on the capillary-flow-induced wire alignment within a sub-micron channel network. In the second approach, electrostatic interaction will be used to selectively position molecular wires on a substrate patterned with different surface charges. Finally, variable temperature electron transport measurement will be carried out on the  $[\text{Mo}_3\text{Se}_3]_\infty$ /surfactant mesostructures and their device configurations. The effect of coupling strength and wire configuration on their transport properties will be addressed.

## Accomplishments

Integration of nanowire and molecular wire building blocks into complex functional structure in a predictable and controlled way represents a major scientific challenge in the nanowire research community. We have developed a simple and parallel method dubbed the microfluidic assisted nanowire integration (MANI) process. The microchannels are formed between a poly(dimethylsiloxane) (PDMS) micro mold and a flat silicon/glass substrate. The microchannels have variable height of 1 to 4 microns, width of 1 to 10 microns and length of 5 to 10 mm. This technique has been successfully applied for the alignment of  $\text{Mo}_3\text{Se}_3^-$  molecular wires, conducting polymer nanowires and carbon nanotubes. Take the  $\text{Mo}_3\text{Se}_3^-$  molecular wires as an example, a droplet of the wire solution/suspension was placed at the open end of the microchannels; the liquid filled the channels under capillary effect. After the evaporation of the solvent and the lift-off of the PDMS mold, bundles of molecular wires

(10 to 100 nanometers in diameter) were aligned along the edges of the microchannels and formed a parallel array. After patterning the first layer of nanowires, the process can be repeated to deposit multilayer nanowires and form complex structures. By rotating the microchannel  $90^\circ$  during the second application, we are able to fabricate arrays of nanowire cross-junctions in a well-controlled and reproducible fashion. This method provides a general and rational approach for the hierarchical assembly of one-dimensional nanomaterials into well-defined functional networks.

In addition, we have also developed a surfactant-induced assembly process for the organization of molecular chains of  $\text{Mo}_3\text{Se}_3^-$ . These infinite chains undergo mesoscopic organization when exposed to opposite-charged surfactants. Low angle x-ray diffraction and transmission electron microscopy studies indicate that the inter-chain spacing can be varied from 20 to 40 angstroms, depending on the alkane length of the surfactants, while the crystallinity along the chains is maintained. This process has also been used to exchange the cations from lithium to sodium, rubidium, cesium, and thallium. This capability of tuning the inter-molecular-wire spacing should enable us to tune the wire-wire coupling strength at the molecular level and hence their electronic properties. In fact, variable temperature I-V measurement indicates that molecular wire bundles with different inter-wire spacing and cations exhibit metallic, semiconducting, and superconducting properties. For example, superconductivity was detected in  $\text{TiMo}_3\text{Se}_3$  nanowires prepared using this solution methodology. This is the first example of tuning nanowire properties through variation of coupling strength among individual molecular wire components.

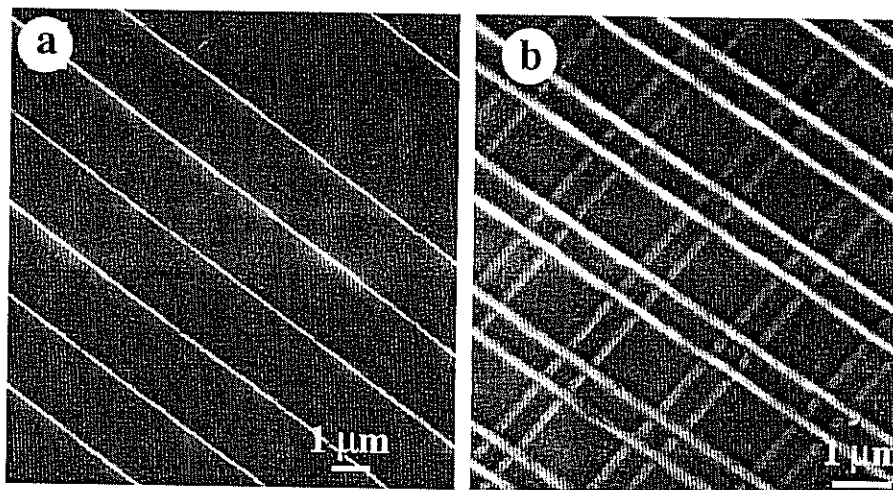


Figure 33: Scanning electron microscopy images of patterned  $\text{Mo}_3\text{Se}_3$  nanowires using MANI process.

## Publications

- J. Song, Y. Wu, B. Messer, and P. Yang, " $\text{MMo}_3\text{Se}_3$  ( $\text{M}=\text{Li}^+$ ,  $\text{Na}^+$ ,  $\text{Rb}^+$ ,  $\text{Cs}^+$ ,  $\text{NMe}_4^+$ ) Nanowire Formation via *Chimie Douce* Reaction," *Journal of the American Chemical Society*, **123**, 9714 (2001).
- J. Song, Y. Wu, B. Messer, and P. Yang, "Redox Templating Synthesis of Metal Nanowires," *Journal of the American Chemical Society*, **123**, 10397 (2001).
- B. Messer, J. Song, and P. Yang, "Microchannel Networks for Nanowire Patterning," *Journal of the American Chemical Society*, **122**, 10232 (2000).
- B. Messer, M. Huang, J. Song, and P. Yang, "Surfactant Induced Mesoscopic Organization of Inorganic Molecular Wires," *Advanced Materials*, **12**, 1526 (2000).





## Nuclear Science Division

---

### ***Design of Digital Signal Processing Electronics for High-Resolution Solid-State Detectors***

---

Principal Investigators: I-Yang Lee and Kai Vetter

Project No.: 01032

#### **Project Description**

The goal of this project is to design and build a prototype of a general-purpose digital signal processing (DSP) system for use with the next-generation of radiation detectors, such as Gamma-Ray Energy Tracking Array (Greta). These new detectors require signal-processing capabilities that can only be achieved with DSPs. The proposed DSP system has the potential to greatly improve the capabilities of many types of radiation detectors. Examples include highly segmented or pixilated solid-state detectors, such as Ge, Si or CdZnTe, that play a vital role not only in basic research (nuclear, astrophysics etc), but also in many applied areas, such as medical imaging and national safety (particularly regarding the transport and storage of radioactive material). In addition, this DSP system will also provide important proof-of-principle for other future experiments.

The design of this DSP system will enable energy, time, and position information of, for example, gamma-ray interactions to be obtained, even at very high event rates for many channels (30 to 40) on one board. Each DSP channel will consist of three stages: (1) a signal filter stage, (2) a digitizer stage, and (3) a processing stage. First, we will determine the physics requirements of the data processing, and then, derive the parameters of the electronics and processing software. Finally, a prototype will be constructed and tested with actual detectors. This project will use the unique capability and experience at Berkeley Lab to design, build, and test a prototype of a multi-channel DSP system, and will draw upon the scientific and technical expertise in this area that is available within the Nuclear Science, Engineering, and Physics Divisions.

#### **Accomplishments**

During the first year of the project, the specification and electrical design for the Greta preprocessor board was completed. The specification requires a multi-channel 12-bit flash analog-to-digital converter (ADC) board with a

sampling rate of 100 MHz. Flexible triggering and programmability were high priorities. This specification was endorsed by the Greta steering committee as well as the Digital Electronics Working Group from the low-energy nuclear structure physics community.

This 40-channel board employs Analog Devices AD8032 12-bit, 100 MHz flash ADC's. These 40 channels are grouped into 5 blocks of 8 with each block controlled by a common Field-Programmable Gate Array (FPGA). These 5 FPGAs are responsible for routing the full data stream from each ADC to a local memory, generate energy and time information for each pulse, forward local trigger information to a board controller, and respond to board controller requests for readout of the local memories. The board controller FPGA generates prompt master triggers and copies relevant contents of local memories to an output buffer. Communication between the board controller and the block controllers is through an 8-bit differential bus clocked at 400 MHz, giving the preprocessor board outstanding bandwidth. The output buffer can be read through a fast optical link or by a board mounted central processing unit (CPU) that has a 10/100 BaseT network for diagnostics.

Several critical elements of this board were successfully tested in the last year. The front-end analog stage of the board was prototyped and meets the effective resolution requirements for the Greta detector. Also, logic design for the block controller FPGA / memory interface was completed and successfully simulated as was the board controller FPGA / local CPU interface. Layout of the circuit board for an 8-channel prototype board and parts procurement were also completed.

This design was presented at several workshops and met with enthusiastic response.

#### **Publications**

K. Vetter, "Electronics for Gamma-Ray Tracking," Workshop on Gamma-Ray Tracking Detectors for Nuclear Science, University of Massachusetts Lowell (June 22-23, 2001).

K. Vetter, "Digital Processing of Signals from Segmented Ge Detectors," Workshop on Experimental Facilities for Isotope Separator and Accelerator Facility (at Vancouver Canada) Phase II (ISAC-II), Lake Louise, Alberta, Canada (February 15-16, 2001).

J. Ludvig, "A General Purpose Waveform Recording DAQ System for High-Energy and Nuclear Physics," Workshop

on Digital Electronics for Nuclear Structure Physics, Argonne National Laboratory (March 2-3, 2001).

K. Vetter, I.Y. Lee, M. Maier, and D. Radford, "Report on Workshop on 'Digital Electronics for Nuclear Structure Physics'," Workshop on Digital Electronics for Nuclear Structure Physics, Argonne National Laboratory (March 2-3, 2001).

---

## ***First Chemical Study of Element 108, Hassium***

---

Principal Investigators: Heino Nitsche and Uwe Kirbach

Project No.: 99033

### **Project Description**

The purpose of this project is to develop the prerequisites for and carry out a first-time study of the chemical properties of hassium (Hs), element 108. The major goals of the program are:

- Development, construction, and testing of the Recoil product Transfer Chamber (RTC) interfaced to the Berkeley Gas-filled Separator (BGS) and a chemical reaction unit where the hassium undergoes chemical conversion.
- Design and testing of a Cryo-Thermochromatographic Separator (CTS). The CTS thermochromatographically separates the hassium reaction products from other actinide reaction products and identifies them by  $\alpha$  decay.
- Study for the first time the chemical properties of hassium and determine if it behaves in a similar manner as its chemical homologues ruthenium (Ru) and osmium (Os).

The chemical identification will be achieved by combining a novel chemical separation system with the advantages of the BGS. The BGS provides good and fast separation of transfer products stemming from impurities of the nuclear target and from long-lived spontaneous fission products. The novel chemical separator is based on the expected high volatility of the tetroxides of hassium and consists of a chemical reaction chamber and the CTS.

The development of the RTC is an important part of this project and is needed to directly couple the BGS with the

chemical separation system, which will facilitate further separation from the actinides and detection by nuclear counting. In the chemical reaction chamber, the nuclear reaction products coming from the RTC via a gas jet are subjected to an oxygen/helium gas mixture at a reaction temperature of 1200°K to form volatile hassium tetroxides. The volatile oxides are then transported via the gas stream through a capillary to the CTS, leaving behind non- and less-volatile compounds. The CTS consists of an assembly of two rows of 32 silicon positive-intrinsic-negative (PIN)-diodes arranged opposite each other, forming a narrow rectangular channel through which the transport gas flows. A negative temperature gradient ranging from room temperature to about 150°K is applied to the PIN-diode assembly. This results in the deposition of the hassium tetroxide on one of the detectors, where it can be identified by  $\alpha$ -counting. We expect a separation factor of  $10^7$  to  $10^9$  for actinides from the combined BGS-CTS system.

For the production of hassium, which has an estimated half-life of about 9.3 seconds, we will use the reaction  $^{248}\text{Cm} + ^{26}\text{Mg} \rightarrow ^{269}\text{Hs} + 5n$ . As model experiments, the chemical volatility and the chromatographic properties of the oxides of the lighter hassium-homologue osmium will be studied in on-line experiments using short-lived  $\alpha$ -decaying isotopes.

### **Accomplishments**

This was the last year of this successful three-year project. In FY 1999, we designed and built the Recoil product Transfer Chamber (RTC), the transfer device between the BGS and a chemical separation system. In FY 2000, the CTS was designed as a separation and  $\alpha$ -decay detection system for the highly volatile tetroxides of osmium and hassium, element 108. Details of the RTC and CTS are given in last year's report.

In FY 2001, with the help of the novel Cryo-Thermochromatographic Separator (CTS) technique, developed as part of this research program, and together with an international team of scientists, we performed the first chemical studies of element 108, hassium, the heaviest element whose chemistry has yet been studied. Full-scale experiments using the CTS technique were performed on osmium oxide at Berkeley Lab's 88-Inch Cyclotron (see below), but the necessary weeks of beam time for the hassium experiments were only available in Europe.

The team established that hassium forms a gaseous oxide similar to that of osmium, confirming that hassium, like osmium, is a member of group 8 of the periodic table and should be placed directly under it.

We helped colleagues at the Paul Scherrer Institute and the University of Bern in Switzerland build a version of the Berkeley Lab CTS detector, which was installed at the Universal Linear Accelerator (UNILAC) at the Gesellschaft für Schwerionenforschung (GSI) in Darmstadt, Germany.

(Hassium was discovered at GSI in 1984; its name comes from Hassias, Latin for Hesse, the state where Darmstadt is located.)

At GSI's UNILAC, magnesium-26 projectiles bombarded targets of curium-248, prepared for the experiment at the Institute of Nuclear Chemistry in Mainz, Germany. Multiple curium targets were mounted in a rotating wheel system developed at GSI. The hassium atoms formed by impacts between target and beam reacted with oxygen to form hassium oxide molecules. The single molecules were carried through the detector by a stream of helium and immediately condensed on semiconductor diodes, arranged in rows and maintained at temperatures graded from minus 20 to minus 170 degrees Centigrade.

The 192.7-MeV beam energy of  $^{26}\text{Mg}^{5+}$  from the accelerator resulted in projectile energies in the  $^{248}\text{Cm}$  targets of 144.0 to 146.5 MeV. The nuclides  $^{270}\text{Hs}$  and  $^{269}\text{Hs}$  were produced via the  $^{248}\text{Cm}(^{26}\text{Mg}, 4n)$  and  $^{248}\text{Cm}(^{26}\text{Mg}, 5n)$  reactions. One four-member and four three-member decay chains were observed. Three decay chains were attributed to the decay of  $^{269}\text{Hs}$  and two decay chains were tentatively attributed to the decay of the new nuclide  $^{270}\text{Hs}$ .

Hassium oxide was found to condense at a higher temperature than osmium oxide, indicating that it is less volatile. The successful chemical separation of hassium points the way to techniques that will be useful in studying the chemistry of other recently discovered elements.

In addition to Nitsche and Kirbach, members of the Berkeley Lab/University of California, Berkeley, team that participated in the chemical studies of element 108 include Cody Folden, Tom Ginter, Ken Gregorich, Diana Lee, Victor Ninov, Jon Petter Omtvedt of the University of Oslo, Joshua Patin, Nicole K. Seward of the University of Surrey, Dan Strellis, Ralf Sudowe, Philip Wilk, Peter Zielinski, and Darleane Hoffman.

## Publications

U.W. Kirbach and C.M. Folden III, *et al.*, "The Cryo-Thermochromatographic Separator (CTS): A New Rapid Separation and  $\alpha$ -Detection System for On-line chemical Studies of Highly Volatile Osmium and Hassium ( $Z=108$ ) Tetroxides, *Nuclear Instruments & Methods in Physics Research, Section A* (in press).

A. Türler and Ch.E. Düllman, *et al.*, "Evidence for the New Nuclide  $^{270}\text{Hs}$ ," submitted to *Physical Review Letters*.

H. Nitsche and U.W. Kirbach, *et al.*, "Toward On-line Chemistry Studies of Hassium (element 108) at the Berkeley Gas-Filled Separator," presented at Heavy Element Research 2 Symposium, Actinides 2001

International Conference, Hayama, Japan (November 4-9, 2001) (LBNL-47858 Abs.).

U.W. Kirbach and H. Nitsche, *et al.*, "Heavy Element Chemistry Studies at the Berkeley Gas-Filled Separator," presented at 221<sup>st</sup> American Chemical Society National Meeting, San Diego, California (April 1-5, 2001) (LBNL-47182 Abs.).

U.W. Kirbach and K.E. Gregorich, *et al.*, "A New Interface for Heavy Elements Studies at the Berkeley Gas-Filled Separator," presented at 221<sup>st</sup> American Chemical Society National Meeting, San Diego, California (April 1-5, 2001) (LBNL-47181 Abs.).

U.W. Kirbach and H. Nitsche, *et al.*, "Superheavy Element Chemistry Studies at the Berkeley Gas-Filled Separator," oral and poster presentation at the Heavy Element Chemistry Contractors' Meeting, Argonne National Laboratory, Chicago, Illinois (November 19-21, 2000).

U.W. Kirbach and K.E. Gregorich, *et al.*, "A New Interface for Heavy Element Studies at the Berkeley Gas-Filled Separator," presented at Chemical and Nuclear Properties of the Heaviest Element: A Symposium in Memory of Glenn T. Seaborg, Division of Nuclear Chemistry & Technology, 220<sup>th</sup> American Chemical Society National Meeting, Washington, D.C. (August 20-24, 2000).



# Physical Biosciences Division

---

## ***SCOR: A Structural/Functional Classification of RNA***

---

Principal Investigators: Steven Brenner and Stephen Holbrook

Project No.: 01033

### **Project Description**

The specific aim of this project is to construct a structural and functional classification of RNA molecules. This includes collating and studying all of the known RNA structures; validating classification plans; constructing a computer database system and public interface for the multipart, hierarchical, and interlinked classifications; and organizing the known structures into the system. We embark on this project with four primary goals: (1) provide a valuable resource to the growing community of biologists interested in RNA structure, in analogy to the Structural Classification of Proteins (SCOP) database of protein structure; (2) discover new features of RNA structure, and especially relationships to function, from the comprehensive organization; (3) lay the groundwork for developing methods of detection of RNA in sequence (especially genomes) from the classified structures; and (4) enumerate and classify substructures for model building and RNA engineering.

Our approach will be to explore the complete repertoire of known RNA structures, and from them learn the most appropriate classification method. Currently, we expect to have six different types of classifications: biological/functional; structural domains; structural motifs; functional motifs; base pairing; and base stacking. The structural domains and biological/functional classifications will be hierarchical, and all the different classification approaches will be interlinked—as it is from these links that we expect to learn about the relationships between the function and structure, and from local to global features. Initially, the database will be populated through manual inspection and the database system itself will be computationally implemented. In addition, energetic and stereochemical calculations will be done in collaboration with others. We intend to use the database as a basis for applying computational methods to recognizing RNA molecules based on sequence.

### **Accomplishments**

In the year of this funding, we made significant progress in constructing several classifications of RNA molecules and their structures. While none is yet complete, each has yielded immediate discoveries. Together, they comprise the foundation of several future research directions. A public version of the database has been released at <http://scor.lbl.gov/>, and a paper describing the initial work has been published.

#### *Evolutionary Classification of Biological RNAs*

The highest level of RNA classification is a summary of known naturally occurring RNAs grouped by biological function. Only a fraction of known RNAs have been structurally characterized, but the information available is already significant and growing rapidly. In addition to structural references, our database also stores functional, sequence, and evolutionary data for the known RNAs. We expect that soon most biologists will require an evolutionary classification database to keep track of all available information. The goal of this level of classification is to relate function to sequence and structure, determine evolutionary relationships, and to assign novel RNAs to known RNA types.

#### *Structural Classification of Domains and Folds*

Large biological RNAs can often be divided into spatially distinct structural subunits or domains that in turn can be partitioned into simpler subdomains or folds. These structural domains, such as the P4-P6 domain of the group I intron, have been experimentally shown to fold into the same structure as found in the complete RNA. Identification and classification of these domains and folds and characterization of their sequence-structure relationships will clarify their roles in biological systems. We have written a program that searches for domains in large RNAs (or proteins) as continuous sequences of high density (residues/volume). We have identified domain boundaries in ribosomal RNAs that correspond to previous physical and biological studies. Within these large domains, simple structural folds can be identified at higher density levels. We have determined simple folds such as transfer RNA (tRNA) and the hammerhead and hepatitis delta virus (HDV) ribozymes to have similar densities. Our goal is to identify and classify common RNA folds within the ribosomal RNA and other larger RNAs.

## Structural Motifs

RNA motifs are the fundamental building blocks of RNA structure. They can be recognized and characterized by their primary, secondary, and tertiary structure conservation. For the motif classification, a total of 309 Protein Data Bank (PDB) entries were examined; this includes all RNA-containing entries released before August 10, 2000, arising from 242 primary references. Figure 34 shows that almost half of these structures are Watson-Crick (WC) duplexes or duplexes with external loops or internal loops only.

“Internal loops” were defined as regions within WC duplexes of one or more nucleotides that do not form WC base pairs (i.e. they are either unpaired or in non-canonical pairs). While most of these have an easily described structure, such as a series of stacked, non-canonical base pairs, a simple dinucleotide platform or a platform as part of a base triple, there are nine complex loops of intricate structure. One intriguing discovery is a U-like turn in a highly asymmetric internal loop, 1AM0 in which G8, A9, A10 and an adenosine-5-monophosphate (AMP) ligand

form a classic GNRA loop (closed by the non-WC G7 - G11 pair). We have also identified two novel classes of internal loop motifs containing base triples. The first of these involves a looped-out base that forms a major groove triple with the second subsequent 3' base pair (N+2). The second motif has a looped-out base forming a minor groove triple with the 5' preceding base pair (N-1). The global structural effect and the functional role of these motifs are yet to be explored.

“External loops” were defined as a series of nucleotides, not WC base paired, on a single strand flanked by a WC base pair. The most striking feature of external loops is that they cluster into a small number of classes: 45% are U-turns, and a similar fraction is classified as tetraloops. At the intersection of these two, 28 loops have the GNRA U-turn tetraloop motif. We have also identified and classified a previously unnamed motif (which we dub “protein binding tetraloop”) with the sequence patterns AYNR or GGNG. These comprise nearly 10% of the external loop structures. An intriguing discovery was that the C and G residues of the evolutionarily identified CUUG tetraloop in 1RNG are WC paired, yielding a structural diloop.

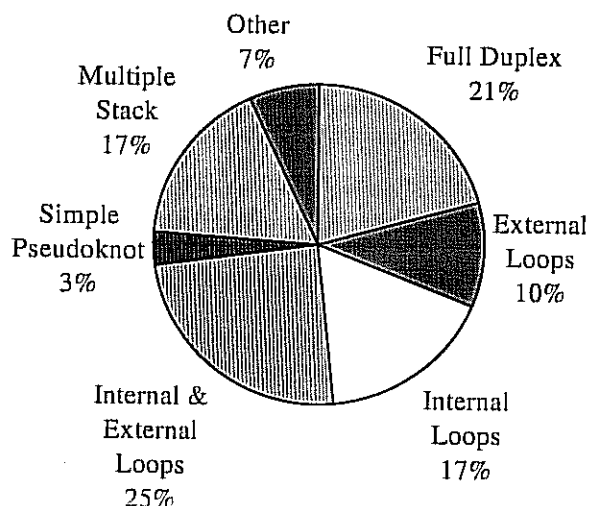


Figure 34: RNA Structures in the Protein Data Bank (PDB).

## Publications

P.S. Klosterman, M. Tamura, S.R. Holbrook, and S.E. Brenner, “SCOR: A Structural Classification of RNA Database,” *Nucleic Acids Research* **8**, 392-394 (2002)  
<http://scor.lbl.gov>.

M. Tamura and S.R. Holbrook, “Sequence and Structural Conservation of RNA Ribose Zippers,” (in preparation).

P.S. Klosterman, M. Tamura, S.R. Holbrook, and S.E. Brenner, “Regular Features in RNA Three-Dimensional Motifs,” (in preparation).

---

## ***Development and Application of the General Theory of Hydrophobicity to Interpret Stability and Dynamics of Biological Assemblies***

---

Principal Investigators: David Chandler

Project No.: 00024

### **Project Description**

While hydrophobic interactions are of fundamental importance in structural biology, a general theory for these forces has not existed. The primary source of the difficulty is the well known yet puzzling multifaceted nature of hydrophobic interactions. We have shown that the resolution of the difficulty is found in the different roles of hydrophobicity at small and large length scales. We have derived equations that describe these different roles and the competition between them. This development opens the way for a quantitative understanding of microscopic and mesoscopic assembly and adhesion. The purpose of this research is to carry out such extensions and applications of the theory, specifically to systems of biological interest.

Specifically, we will develop generally applicable algorithms to apply the Lum-Chandler-Weeks theory equations. These algorithms will be applied in conjunction with molecular dynamics. This approach, coupled with a course-grained dielectric description of electrostatics, provides an implicit solvent model for biomolecule solvation.

### **Accomplishments**

This and the previous year of Laboratory Directed Research and Development (LDRD) funding allowed my research group to carry out several applications and extensions of our recently developed theory of hydrophobicity.

First, we used the theory to successfully interpret the temperature dependence of protein folding. The Lum-Chandler-Weeks theory of hydrophobicity was applied to treat the temperature dependence of hydrophobic solvation in water. The application illustrates how the temperature dependence for hydrophobic surfaces extending less than 1 nanometer differs significantly from that for surfaces extending more than 1 nanometer. The latter is the result of water depletion, a collective effect, that appears at length scales of 1 nanometer and larger. Because of the contrasting behaviors at small and large length scales, hydrophobicity,

by itself, can explain the variable behavior of entropies of protein folding.

Second, we have been able to interpret the interfacial structure of water near an oily surface, and to analyze the length scale dependence of hydrophobic interactions. For example, we have calculated the free energy of solvation for hard sphere solutes, as large as 20 angstroms in diameter, in two simple-point-charge models of water. These results were obtained using umbrella sampling of ensembles with fixed, ambient temperature and pressure. For the same water models, we have also calculated the surface tension of a liquid-vapor interface at room temperature.

We have also tested our theory and alternatives against results of molecular simulation and demonstrated that our theory is quantitatively successful. In one case, we studied the spatial density of a molecular fluid in the presence of a solute of arbitrary size and shape. The density functional was written in a way that effectively describes small deviations around the uniform density, plus an energy density part that is responsible for formation of liquid-vapor interface. Using the weighted density approach, we required the density functional to match with several observed properties of the fluid, such as equation of state and surface tension. We also showed that weighting functions for calculating the weighted density could be obtained from experimental data. Using these elements, we constructed a spatial density functional theory of water and applied it to obtain densities and solvation energies of a hard-sphere solute with encouraging results.

In still another study, we formulated our theory in a fashion that allows it to be combined usefully with computer simulation of biopolymers and other large assemblies. Here, we presented a statistical field theory to describe large length scale effects induced by solutes in a cold and otherwise placid liquid. We found that additional terms in the Hamiltonian can provoke large length scale effects, such as the formation of interfaces and depletion layers. We applied our theory to compute the reversible work to form a bubble in liquid water, as a function of the bubble radius. Comparison with molecular simulation results for the same function indicates that the theory is reasonably accurate. Importantly, simulating the large length scale field involves binary arithmetic only. It thus provides a computationally convenient scheme to incorporate explicit solvent dynamics and structure in simulation studies of large molecular assemblies.

Finally, we have used this formulation to study the dynamics of the folding of a small polymer, showing the relative importance and differing roles of solvent depletion forces and solvent drying.

## Publications

D.M. Huang and D. Chandler, "The Hydrophobic Effect and the Influence of Solute-Solvent Attractions," submitted to *Journal of Physical Chemistry*.

D.M. Huang, P.L. Geissler, and D. Chandler, "Scaling of Hydrophobic Free Energies," *Journal of Physical Chemistry*, **105**, 6704 (May 10, 2001)

S.X. Sun, "A Weighted Density Functional Theory of Solvophobic Effect," *Physical Review E*, **64**, 021512 (July 24, 2001)

P.R. tenWolde, S.X. Sun, and D. Chandler, "Model of a Fluid at Small and Large Length Scales and the Hydrophobic Effect," to appear in *Physical Review E*.

P. R. tenWolde and D. Chandler, manuscript in preparation.

---

## Molecular Recognition and Protein/Protein Interactions in Signal Transduction

---

Principal Investigators: Thomas Earnest

Project No.: 01034

## Project Description

The interactions of proteins with other proteins, RNA, or DNA give rise to a great number of molecular and cellular functions. As genomic sequencing projects produce large numbers of samples, the need to characterize these samples in terms of their function becomes of great importance. We will use members of the Wnt signaling pathway—important in development and cancer—to explore interaction partners using molecular and cellular biology, along with synchrotron-based biological crystallography of the molecular complexes. We will also initiate strategies and methods to automate the exploration, expression, and structural studies of functionally interacting biomolecules.

In particular, we will use two-hybrid analysis to systematically investigate and identify the interactions of proteins in the Wnt pathway with other proteins or sets of proteins. Once these interacting partners are identified, co-

expression in a variety of expression systems, followed by purification, crystallization, and structure determination of the complexes will proceed. Three-hybrid screens will be attempted once binary complexes are identified. The ability to discover, express, and crystallize higher-order complexes will be pursued for structural studies at the Berkeley Center for Structural Biology (BCSB) at the Advanced Light Source. Verification of interactions will proceed by molecular biological, biochemical, structural, and cellular methods. Development of robotic methods and approaches for high-throughput protein-protein interaction studies will be a high priority.

## Accomplishments

Over the past year we have studied molecular recognition events between proteins, focused primarily on proteins of the Wnt signal transduction pathway, which is involved in several developmental processes. Included in this research is the development of methods and technology to improve our ability to study these interactions, in particular by x-ray diffraction to determine the three-dimensional structure of proteins and protein complexes. Our collaboration with Randy Moon, Howard Hughes Medical Institute and University of Washington) has led to the structure of the disheveled PDZ domain in complex with carboxy-terminal peptides from Dapper, a Wnt signaling antagonist identified from a two-hybrid screen using the disheveled PDZ domain as bait. These complexes demonstrate the mode of disheveled/Dapper interactions and have been combined with information from a number of cellular assays to describe a model for Dapper and disheveled in neural development.

We have also obtained high-level expression and purification of a number of other members of the Wnt pathway: beta-catenin (Arm 1-Cterminal), CBP (KIX domain and ~ 100 preceding residues), CtBP (full-length), and CtBP/TCF. These are currently being characterized individually and with appropriate partner proteins. Crystallization trials are ongoing.

Since several constructs of each protein need to be screened for expression and solubility before purification and crystallization trials can proceed, we have also developed a flowchart for increasing the throughput of this process. This also has led to the development of automation for this process, which requires large numbers of samples, but at small volumes for the initial screen. We are currently using green fluorescent protein as a visible reporter for expression and solubility.



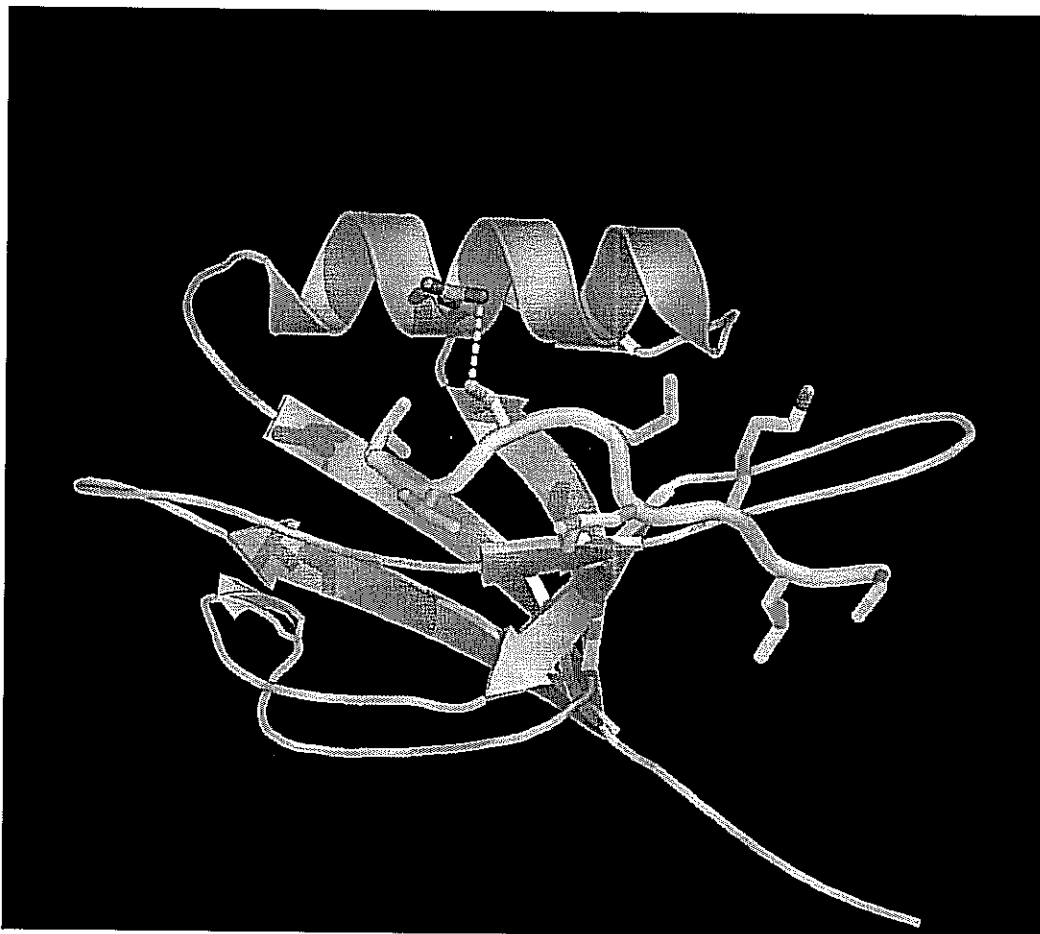


Figure 35: Complex of *Xenopus* disheveled PDZ domain with the carboxy-terminal of *Dapper*, a Wnt signaling antagonist.

## Publications

B.N.R. Cheyette, J.S. Waxman, J.R. Miller, J.M. Seeling, N. Khlebtsova, K. Takemaru, L.C. Sheldahl, E.P. Fox, D.M. Virshup, T.N. Earnest, and R.T. Moon, "Dapper, a Dishevelled-Associated Antagonist of Wnt Signaling, is Required for Notochord and Head Formation," reviewed and submitted to *Cell*.

N. Khlebtsova, L-W. Hung, B.N.R. Cheyette, J.R. Miller, R.T. Moon, and T.N. Earnest, "Conformational Flexibility in the PDZ Domain of Dishevelled Protein Induced by Target Binding," in preparation for *Molecular Cell*.

---

## Teraflop Challenges in Single-Particle Electron Crystallography

---

Principal Investigators: Robert Glaeser, Kenneth Downing, Eva Nogales, Esmond Ng, and Ravi Malladi

Project No.: 00017

## Project Description

The goal of this project is to define efficient mathematical approaches (and algorithms) that produce a three-dimensional reconstruction from high resolution, cryo-

electron microscopy (EM) images of single protein molecules. These tools must make it possible to automatically identify between  $10^5$  and  $10^6$  single particles and automatically merge these images to produce the three-dimensional reconstruction. In order for this to be practical, identification and merging should be accomplished with less than  $\sim 10^{17}$  floating point operations ( $\sim 10^5$  teraflop). The long-term goal is to be able to carry out structural studies of large, multi-subunit protein complexes at high resolution, using electron microscope images of fields that contain  $\sim 100$  particles each. Merging data from single particles is equivalent to crystallization *in silico*. By eliminating the need for biochemical crystallization, and by reducing data collection and three-dimensional reconstruction to about one day each, single-particle electron crystallography will achieve a level of high throughput that is similar to the speed of x-ray crystallography. There will be no delay in screening for crystallization conditions, however. Furthermore, structural studies will be made possible for complex molecular assemblies that represent increasingly problematic challenges for x-ray crystallography, but which are increasingly more favorable for cryo-electron microscopy.

To accomplish this, existing methods of automatic particle identification will be refined by the addition of conceptually new mathematical tools. The goal is to improve particle identification to a level such that false negatives fall below 25% and false positives fall below 10%. In parallel with this effort, we will port existing software suites (SPIDER) to the Alvarez computer at the National Energy Research Scientific Computing Center (NERSC), and we will use existing data sets (one with almost  $10^5$  particles) to demonstrate that routine, high-throughput merging of very large data sets is feasible on this machine.

## Accomplishments

Work during 2001 has focused on two problems: (1) development of computational tools that can be used for automated identification of large, macromolecular particles in images that have been recorded with an electron microscope, and (2) development of versions of the SPIDER software that run efficiently on large, distributed memory machines.

In our first step we had conducted a review of the literature in which numerous attempts had been made to automate the task of identifying single particles. This review was submitted for publication, and appeared May 2001. The Malladi group have gone on to demonstrate that a gradient-based mathematical smoothing of cryo-electron microscopy images is effective at preserving the contrast-edge between a particle and the surrounding ice, thereby making it possible to accurately define a contour that surrounds the particle. A major advance has also been made in speeding the execution of the smoothing operation by a factor of at

least 20, with no apparent degradation of preserving the edge. A remaining problem occurs when particles are quite close to one another, but a likely solution will be to modify the stopping criteria applied during active contouring so as to take the sign of the gradient into account.

Significant progress has been made by the Ng group in the design of algorithms for matrix conditioning that can be applied to the task of three-dimensional reconstruction in electron microscopy. This preliminary data provides crucial support for a grant application that seeks National Institutes of Health (NIH) funding of a Program Project, which we wish to establish on the basis of the seed-money provided by Laboratory Directed Research and Development (LDRD) funding. Following completion of the feasibility study on use of sparse matrix techniques, attention was turned to creating a parallelized implementation of some of the most time-intensive operations within SPIDER. This parallelization has been accomplished with the use of the Message Passing Interface (MPI) standard, and tests have been successfully run on 50 nodes of the Alvarez cluster at Berkeley Lab. While much further work remains along this line, including a larger restructuring to adopt a "publish and subscribe" strategy for load balancing, the work accomplished in FY 2001 provided crucial advances in computational aspects of the reconstruction of protein molecules.

## Publications

W.V. Nicholson and R.M. Glaeser, "Automatic Particle Detection in Electron Microscopy," *Journal of Structural Biology* 133, 90-101 (2001).

---

## Cooperative Effects Determining Fidelity in Cellular Recognition

---

Principal Investigators: Jay Groves, Carolyn Bertozzi, and Arup Chakraborty

Project No.: 01035

## Project Description

Cells interact with each other and their environment through myriad membrane-associated receptors and signaling molecules. Dynamic spatial reorganization of membrane receptors is a critical aspect of recognition and signal transduction mechanisms in a number of systems. Activation of T-lymphocytes is a prominent example. Initiation of the immune response requires large-scale, spatial self-assembly of T-cell receptors (TCR) engaged

with major histocompatibility protein (MHC)-peptide complexes on an antigen presenting cell (APC). Formation of such an immunological synapse triggers an immune response. Although the cooperative reorganization of membrane proteins leading to the formation of functional immunological synapses has recently been observed, a physical understanding of the forces driving their organization remains elusive. We plan to mount a quantitative investigation of the physical characteristics and principles governing molecular reorganization events during initial stages of cellular recognition and signaling. Elucidating these principles may also reveal unexpected drug targets as well as biologically inspired strategies for recognition and patterning useful for materials design.

Our approach is aimed toward elucidating how physico-chemical parameters determine the formation of spatio-temporal patterns. This will allow us to delineate the conditions that lead to the formation of signaling junctions. An investigative platform that combines novel membrane experiments and synthetic biochemistry with sophisticated theoretical calculations and computer simulations has been formulated to meet our goals.

## Accomplishments

### *Synaptic Pattern Formation*

The membrane introduces a fluctuating two-dimensional reaction environment in which topographical constraints and restricted component access strongly influence receptor binding and spatial organization. Recently, we have proposed that intrinsic coupling between binding interactions, membrane constraints, and protein mobility can drive spontaneous formation of synaptic patterns within the interaction zone. The differential size of different receptor-ligand pairs is one important cause of coupling between receptor-ligand binding, membrane topography, and large-scale organization. Coupling between the reaction environment and the individual receptor-ligand binding events provides a cooperative signal amplification mechanism that cells may exploit. We have developed a theoretical model incorporating the essential phenomena using statistical mechanical principles. When the parameters that measure receptor-ligand binding kinetics, protein mobilities, and cell membrane characteristics are set according to those measured for the T-cell immunological synapse, the spontaneously evolving patterns are found to be in good agreement with experimental observations. This suggests that synaptic patterns may form spontaneously as a result of a precise balance of forces due to receptor binding, lateral mobility of proteins, and membrane constraints. Active interventions such as those due to directed cytoskeletal flow (in T-cells) are superimposed on these self-organizing tendencies.

Results using our computational model have provided insight into the mechanism by which peptide-mediated

MHC-TCR binding kinetics corresponds to biological activity. An essential function of the TCR is to distinguish between different MHC-bound peptides. Cellular behavior is known to be strongly correlated with the dissociation rate ( $k_{\text{off}}$ ) for the TCR/MHC-peptide complex. Values of  $k_{\text{off}}$  in the range 0.06 to 0.3 s<sup>-1</sup> lead to T-cell activation. Using the spatial pattern of proteins as criteria for effective synapse formation, our calculations show that only MHC-peptide complexes that bind TCR with  $k_{\text{off}}$  in a narrow range (similar to experiments) lead to spontaneous pattern formation with synaptic characteristics. These results indicate that differences in  $k_{\text{off}}$  are amplified by the cell membrane environment to affect biological outcome.

### *Supported Membrane Junction*

We have initiated development of an inter-membrane junction between a supported membrane and a second membrane patch, which is deposited from a giant vesicle. Incorporation of different fluorescent probe lipids into each of the two membranes enables a variety of fluorescence imaging capabilities, which we employ to obtain both lateral and topographical information about the junction. Fluorescence resonance energy transfer (FRET) between the two membranes when they are closely apposed (< 4 nanometers) leaves a footprint, which maps the contact zone. Larger topographical features can be observed by fluorescence interference contrast microscopy (FICM). When the membrane system is supported on an oxidized silicon wafer, near-field interference effects set up optical standing waves that provide contour mapping of the membrane topography.

We are developing convolution data analysis procedures to quantitatively analyze topographical image data collected utilizing the near-field interference effects. Calculated interference patterns are convolved with the impulse-response function of the imaging system and compared to observed data. Using these procedures, we have determined that this method affords us 5-nanometer resolution (in the z-dimension) over a range extending for at least one micron from the surface. The combination of this interference imaging with intermembrane FRET provides an extremely powerful optical system for analysis of three-dimensional intermembrane contact zones.

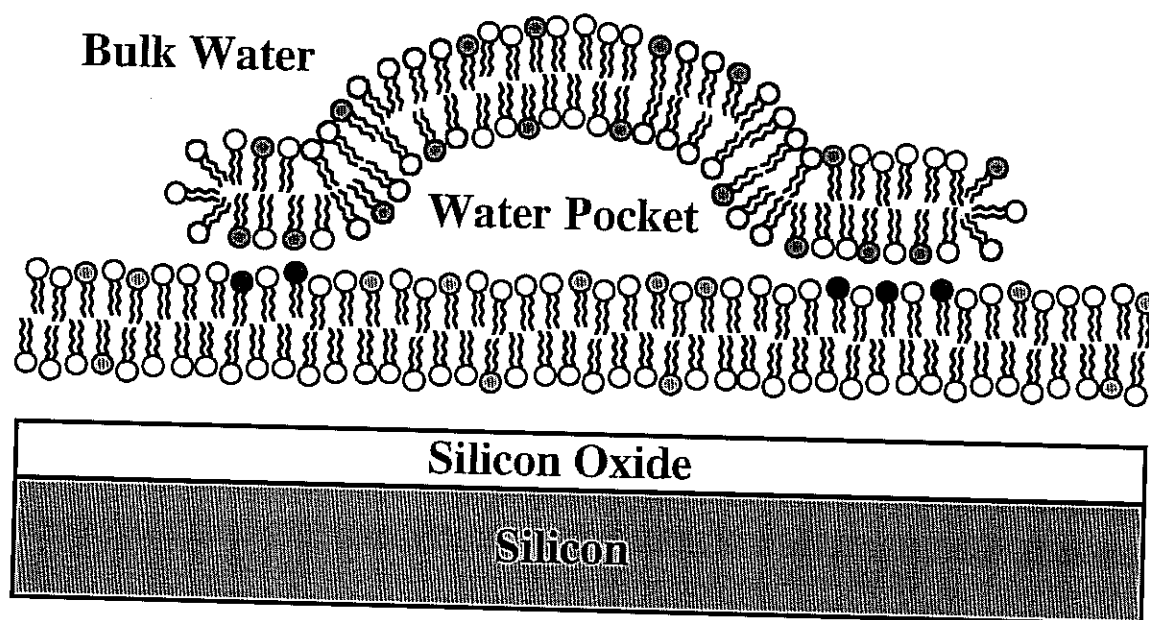


Figure 36: Newly developed supported membrane junction for studies of spontaneous pattern formation in membrane systems.

### Publications

S.Y. Qi, J.T. Groves, and A.K. Chakraborty, "Synaptic Pattern Formation During Cellular Recognition," *Proc. Natl. Acad. Sci. USA*, (June 2001).

A.P.F. Wong and J.T. Groves, "Topographical Imaging of an Intermembrane Junction by Combined Fluorescence Interference and Energy Transfer Microscopies," *J. Am. Chem. Soc.*, (November 2001)

J.T. Groves, L.K. Mahal, and C.R. Bertozzi, "Control of Cell Adhesion and Growth with Micropatterned Supported Lipid Membranes," *Langmuir*, (September 2001).

### Novel Synchrotron Experiments to Determine Hydration Forces for Molten Globules and Model Proteins for Extremophiles

Principal Investigators: Teresa Head-Gordon

Project No.: 01036

### Project Description

The aim of this proposal is to advance the transferability of our solution-scattering/simulation technique to new areas of biological interest, namely that of molten globule proteins, and hydrated, model protein systems that are explored at extremes of pressure and temperature. These new experiments will allow us to address two important biological systems. First is the structural organization of the hydrophobic core of molten globule proteins, a thermodynamic protein-folding intermediate that is thought to be a good model of kinetic intermediates for generic proteins. Second, the same techniques will allow us to explore the changes in physical hydration forces over large ranges in temperature and pressure, to address a

primary research question in the area of extremophile organisms: how proteins necessary for life under these extreme conditions can remain stable and functional.

The experimental repertoire of protein-based structural techniques has resulted in a good understanding of a protein's secondary structure and tertiary structure contacts, while approaches to characterize the role of the hydration environment in terms of structure and forces in folding are comparatively minimal at present. We have combined our expertise in solution-scattering experiments, simulations, and theory, to define an approach to determine hydration structure and forces in protein solutions. We propose to perform novel selenium-edge and second-order difference, solution-scattering experiments at the Advanced Light Source using selenium-methionine derivatives of both amino acid monomers and the protein  $\alpha$ -lactalbumin that will reveal the structural organization of the hydrophobic core. We will also extend these same experiments to understand hydration at extremes of temperature and pressure.

## Accomplishments

This year we have done some preliminary scattering experiments of selenium-based compounds in aqueous solutions to understand the second order difference and selenium-edge experiments and analysis. We are currently analyzing this data.

We have also conducted scattering experiments on pure water over a range of temperature because neutron scattering experiments have proven unreliable. The higher temperature data sets generated by neutron need to be independently confirmed by the oxygen-oxygen radial distribution functions derived from x-ray data, since the problem of inelasticity effects in neutrons are known to increase with increasing temperature. This data has proven very popular with researchers throughout the world, and is reflected in our invited review on water structure in *Chemical Reviews* this year.

## Publications

T. Head-Gordon and J. Wooley, "Computational Challenges in Structural and Functional Genomics," *IBM Systems Journal: Deep Computing in the Life Sciences*, **40**, 265-296 (2001).

T. Head-Gordon, G. Hura, J. Sorenson, and R.M. Glaeser, "Pure Water Structure and Hydration Forces for Protein Folding," *NATO Advanced Research Workshop: New Kinds of Phase Transitions: Transformations in Disordered Substances* (in press).

T. Head-Gordon and G. Hura, "Water Structure from Scattering Experiments and Simulation," submitted to *Chemical Reviews*.

G. Hura, J. Sorenson, R.M. Glaeser, and T. Head-Gordon, "A High-Quality X-Ray Scattering Experiment on Liquid Water at Ambient Conditions," *Journal of Chemical Physics* **113**, 9140-9148 (2000).

J. Sorenson, G. Hura, R.M. Glaeser, and T. Head-Gordon, "What Can X-Ray Scattering Tell Us about the Radial Distribution Functions of Water?" *Journal of Chemical Physics* **113**, 9140-9161 (2000).

---

## Genetically Encoded Optical Sensors of Cell Signaling

---

Principal Investigators: Ehud Isacoff

Project No.: 00025

## Project Description

Fluorescent indicator dyes have revolutionized our understanding of cellular signaling by providing continuous measurements of physiological events in single cells, and also in cell populations with high temporal and spatial resolution. Until now, these organic dyes have been limited to detecting changes in membrane voltage or in concentration of small diffusible regulators or metabolites, such as protons and calcium ions. A second limitation is that these dyes must be synthesized chemically and introduced as hydrolyzable esters or by microinjection, meaning that they either fill all cells in a tissue entirely, or that only one or a small number of cells are labeled. In absence of localization to specific cell types, and specific sub-compartments of cells, optical signals of interest are often drowned out either by indicator background from inactive cells and inactivate parts of cells, or by other signals.

To increase our understanding of the ligand and contact mediated membrane signals that govern development, sensory transduction, and learning and memory it is necessary to greatly expand the range of signaling events that can be detected and to develop means of targeting sensors to specific cellular locations. One general approach to this problem is to make sensors from proteins—the very biological molecules that transduce and transmit cellular signals. This approach would harnesses the high sensitivity and specificity of biological systems, which can detect an enormous range of signals,

in very selective ways, down to single molecule events. Moreover, it could permit the detection of signaling events all the way from the earliest stage of membrane transduction, through the network of signaling relays and amplification steps, and finally to downstream events that occur anywhere from the nucleus back to the plasma membrane.

Since protein-based sensors are encoded in DNA they can be placed under the control of cell-specific promoters, introduced *in vivo* or *in vitro* using gene transfer techniques, and even targeted to specific subcellular compartments using protein signals recognized by the cell's native protein sorting machinery. In addition to solving the problem of sensor production and targeting, DNA-encoded sensors have the advantage that, unlike organic dyes, they can be rationally "tuned" by modification of their functional domains with mutations that are known to adjust their dynamic range of operation.

## Accomplishments

### Strategies for Sensor Design

We have made several new optical sensors for measuring cellular signaling events in living cells. These sensors are based on the idea that the most sensitive and precise reporters of biologically relevant signals, and the only ones that can be assured to be in the right place at the right time to observe interesting events, are the very proteins that transduce and propagate those signals. We have taken the approach of modifying signaling proteins in such a way that changes in their biological activity is converted into changes in fluorescence emission. The sensor protein thus has two parts: the "detector" signaling protein (for example an ion channel, membrane receptor, enzyme, ligand-binding peptide, G-protein, etc.) that undergoes a conformational rearrangement during a cell signaling event, and a "reporter" fluorophore that is attached to the detector at a location that makes its emission sensitive to the detector's conformation.

### Green Fluorescent Protein as a Reporter of Detector Protein Movement

Although attractive for the variety, high environmental sensitivity, and flexibility of attachment site of the small organic dye method, this approach requires a step of labeling before use. For this reason the Isacoff lab has explored an alternative approach in which both the detector and reporter are protein-based, thus avoiding the need for exogenous labeling. This approach employs the Green Fluorescent Protein (GFP) as a reporter. GFP is fused to the detector protein near a domain that undergoes a conformational rearrangement when the detector protein is activated. Detector protein movement either alters the environment of GFP or puts torque on the GFP's structure, thus altering its optical properties. Several years

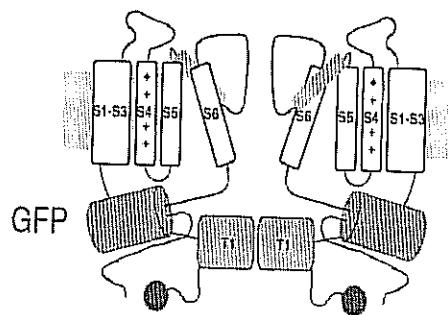
ago, we employed such a strategy by fusing a conformationally-sensitized version of GFP into a voltage dependent channel to make FlaSh, an entirely genetically encoded optical voltage sensor (Figure 37).

### Tuning FlaSh: Redesign of the Dynamics, Voltage Range, and Color of a Genetically-Encoded Optical Sensor of Membrane Potential

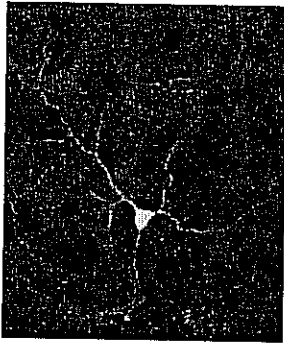
We have now succeeded in tuning the color, kinetics, and dynamic range of FlaSh. We have optimized for the detection of either action potentials (at positive voltages with rapid kinetics), on one hand, or synaptic potentials (at more negative voltages with slower kinetics), on the other. By modifying the GFP reporter, we have produced sensors which all fold efficiently at 37°C (enabling use in mammalian cells), and with distinct spectra. Sensors of different colors can now be targeted to different cell types to enable the synchronous measure of activity from two or three sub-populations of intermingled cells in the same neural circuit, which could not be otherwise distinguished.

We have succeeded in expressing the sensors in three different types of excitable cells (Figure 38). These include two different neuronal cell types from the mammalian brain (hippocampal pyramidal cells and cerebellar granule cells) as well as the pharyngeal muscle of the model genetic organism *C. elegans*.

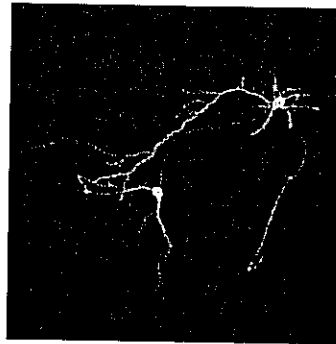
### GFP Insertion into Channel Detector



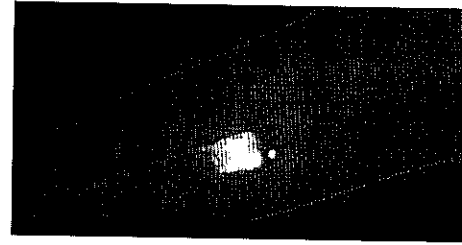
**Figure 37: The FlaSh construct.** Structural model of FlaSh shows two of the four subunits of the Shaker channel. Each subunit contains six transmembrane domains, including the charged voltage sensing S4. The GFP reporter (shaded cylinders) is inserted into the detector channel 25 amino acids away from the internal end of S6—part of the channel's internal gate, and a location where conformational rearrangements in the channel perturb the fluorescence of GFP. The GFP has been sensitized to the rearrangements of the channel by deletion of the last eight residues in the C-terminal, which are disordered in the crystal structure. T1 domains associate into cytoplasmic tetramers and aid in channel assembly. The small dark balls are the N-terminal fast inactivation balls.



**Figure 38A)** Mouse P5 hippocampal



**Figure 38B)** Mouse P5 cerebellar



**Figure 38C)** *C. elegans* pharyngeal muscle

*Figure 38: FlaSh expression in three different types of excitable cells.*

## Publications

G. Guerrero and E.Y. Isacoff, "Genetically Encoded Optical Sensors of Neuronal Activity and Cellular Function," *Current Opinion in Neurobiology*, **11**, 601-607 (2001).

G. Guerrero, M.S. Siegel, B. Roska, E. Loots, and E.Y. Isacoff, "Tuning FlaSh: Redesign of the Dynamics,

Voltage Range and Color of the Genetically-Encoded Optical Sensor of Membrane Potential," in preparation.





## Physics Division

---

### **Modeling of High Energy Physics Detectors**

---

Principal Investigators: Ian Hinchliffe and Murdock Gilchriese

Project No.: 01037

#### **Project Description**

Accurate simulation of the response of high energy physics detectors is essential for the control of systematic uncertainties in the extraction of physics signals. The new object-oriented simulation tool, GEANT-4, has recently become available. Validation of the models underlying this tool is essential and needs to take place well in advance of the tool's actual use. GEANT-4 has application beyond high energy physics. This proposal will undertake a detailed comparison of the predictions of GEANT-4 with a system—the A Toroidal LHC Apparatus (ATLAS) pixel test beam—where data is available. This will enable validation and refinement of the underlying physics models in GEANT-4.

Specifically, detailed comparisons of the predictions of GEANT-4 with actual data are required. Comparison will be made with test-beam data to be obtained at the European Laboratory for Particle Physics (CERN) in the immediate future. A GEANT-4 model of the ATLAS pixel test beam will be made and integrated into the ATLAS software framework. Results from this will be used for a detailed validation of GEANT-4 and to adjust the underlying GEANT-4 model.

#### **Accomplishments**

Led by Berkeley Lab under the direction of David Quarrie, work was undertaken on simulations of the ATLAS detector involving the development of the proposed new ATLAS software framework. The simulations were presented at ATLAS collaboration meetings and have resulted in a publication. A package was developed to enable events from physics generators to provide a uniform interface to the ATLAS simulation package.

Work on integration of GEANT-4 into the new framework has begun and will become the basis of the new full simulation tool being developed for the ATLAS collaboration. This proof of principle has demonstrated that a seamless simulation environment can be established for the ATLAS experiment. Events from the generator's package can be fed into GEANT-4 using this system.

In addition, modeling work on reconstruction of events in ATLAS has progressed. In particular, the package xKalman responsible for inner detector reconstruction (including the pixel system) has been enhanced and made more effective. All of the software produced has been made available to the ATLAS collaboration and forms part of the potential software suite to be tested in the first set of data challenges that will begin in December 2001.

This work has resulted in partial follow-on funding from U.S.-ATLAS project funds [Department of Energy and National Science Foundation (NSF)] being sent to Berkeley in order to continue the work. In particular, a new position is open that is funded by NSF, to take over and extend the integration of the physics generators into the ATLAS framework.

#### **Publications**

I. Hinchliffe and L. Vacavant, "Signals of Models with Large Extra Dimensions," ATLAS report in *Journal of Physics*, G 27, 1839 (March 2001).

---

### ***POLARBEAR: An Experiment to Measure Polarization Anisotropy in the Cosmic Microwave Background***

---

Principal Investigators: Adrian Lee and Helmuth Spieler

Project No.: 01038

#### **Project Description**

This project will conduct research and development in support of a new experiment to measure Cosmic Microwave Background (CMB) polarization. The new experiment, called POLARization of the Background

millimEter bAckground Radiation (POLARBEAR) combines the proven MAXIMA (Millimeter Anisotropy eXperiment Imaging Array) telescope and observation strategies with a new detector technology based on superconducting transition-edge sensors (TES).

The Cosmic Microwave Background is proving to be a peerless laboratory for cosmology and fundamental physics. Recently, measurements of the spatial temperature anisotropy by MAXIMA and BOOMERANG (Balloon Observations of Millimetric Extragalactic Radiation and Geophysics) have given strong support to inflationary models with a density close to critical. We have also been able to measure the density of baryons and cold dark matter, and the spectral index of the primordial power spectrum. Perhaps the most exciting promise of polarization measurements lies in their ability to probe the inflationary epoch, i.e. as early as  $10^{\exp(-38)}$  seconds after the Big Bang. Gravity waves produced at these early times propagate forward and produce structure in the photon-baryon fluid at the surface of last scattering, observable by its signature in the polarization of the CMB. Detection and characterization of this polarization will require a large leap in experimental capability. The key new technology for POLARBEAR is the large-format array of voltage-biased superconducting bolometers that are based on a superconducting film biased in mid-transition. Several tasks will be performed to examine the technical details of such a detector.

## Accomplishments

We have worked on the general design and testing of components for the proposed POLARBEAR receiver. The main thrust areas have been the cryogenic system, the integration of antennas, bandpass filters and sensors, and the readout.

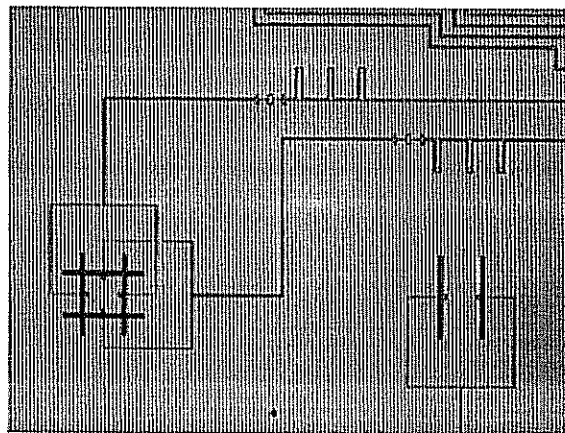
POLARBEAR will use two refrigerators, a "pulse-tube" cryocooler to achieve an intermediate temperature of 2.5°K, and a  $^3\text{He}$  closed-cycle refrigerator to cool the detectors to 300°mK. The use of a cryocooler instead of liquid cryogens will facilitate the long integration times required for the success of this experiment. We have integrated the new cooler in a small test cryostat and have verified a base temperature of 2.3°K, in agreement with specification. We are currently installing a small  $^3\text{He}$  closed-cycle refrigerator and a superconducting bolometer to assess if the detector noise performance is adversely affected by vibrations from the pulse-tube cooler. We have begun the design of the receiver housing. The housing provides a vacuum enclosure and provides radiation shields at 50°K and 2.5°K.

We have recently completed the design for the first antenna-coupled bolometer prototype. Planar antennas

with dual polarization (for coupling to the telescope), microwave band-defining filters, and bolometers are integrated onto a single substrate. We have completed a full electromagnetic simulation of the transmission lines and filters and prepared layout files for fabrication.

We have been investigating the design of an economical Superconducting QUantum Interference Device (SQUID) readout for POLARBEAR. In the long term, we will use a SQUID-based readout multiplexer that we have originated. This multiplexer will enable arrays of several thousand elements. However, before we design and implement such a large array, we will deploy the receiver with a smaller array to test the detectors and observation strategy. The first step will be an ~100-element array that we can build using a commercial SQUID and a custom set of SQUID readout electronics. We have built and tested a prototype of the custom room-temperature SQUID readout. We are doing a design study of the tradeoffs between single AC-modulated SQUIDs and series arrays of SQUIDs.

Finally, we have completed a search for a postdoc, who will join the project in early FY 2002. This will greatly accelerate the exploration of the various options that are being considered.



*Figure 39: Prototype chip of antenna-coupled bolometers. A niobium ground plane and niobium traces over oxide insulator are visible. For scale, the dipole antennas are roughly 1 millimeter in length. The planar antennas are double slot dipoles. Slot dipoles are simply slots cut in the ground plane, and are the Babinet complements of metal dipoles. The antennas accept radiation from the telescope, but suppress radiation from the surroundings. The radiofrequency energy from the antennas propagate along "microstrip" transmission lines, through frequency-band defining resonant filters, and finally is absorbed in the bolometer. The antenna to the left receives two orthogonal polarizations, and the signals from these antennas are differenced after the bolometers to form a differential polarimeter.*

## Publications

M.J. Meyers, A.T. Lee, P.L. Richards, D. Schwan, J.T. Skidmore, A.D. Smith, H.G. Spieler, and J. Yoon, "Antenna-Coupled Arrays of Voltage-Biased Superconducting Bolometers," submitted to *Proceedings of Low-Temperature Detectors (LTD) 2001* (July 23, 2001).

J. Yoon, J. Clarke, J.M. Gildemeister, A.T. Lee, M.J. Meyers, P.L. Richards, J.T. Skidmore, and H.G. Spieler, "Single SQUID Multiplexer for Arrays of Voltage-Biased Superconducting Bolometers," submitted to *Proceedings of Low-Temperature Detectors (LTD) 2001* (July 23, 2001).

---

## Foundations for a SuperNova/ Acceleration Probe (SNAP)

---

Principal Investigators: Michael Levi and Saul Perlmutter

Project No.: 00026, 00027

In FY 2000, Berkeley Lab funded two separate projects. Project 00026 is to tackle technical challenges in developing a large charge-coupled device (CCD)-mosaic camera having near-infrared (IR) sensitivity. Project 00027 is to develop techniques to significantly increase discovery of nearby supernovae—which is particularly constrained by data handling bottlenecks—for reducing the systematic uncertainties in our current understanding of supernova observations. Consequently, the two self-contained reports follow.

For FY 2001, because both scientific problems are crucial to the possible development of the proposed Department of Energy/National Aeronautics and Space Administration SNAP satellite, they were considered effectively a single project. Nonetheless, both scientific problems are crucial for further general advances in astrophysics and astronomy and therefore are of ongoing significance independent of the ultimate success of the SNAP satellite proposal.

---

## Sub-Project 00026: Development of an Advanced Imaging Array

---

### Project Description

A major milestone has been achieved with the recent success in fabricating near-IR sensitive, large-format 2000 by 4000 pixel CCDs. We must now find ways to exploit this technology to the fullest. In particular, these devices can be placed in advanced cameras for observational astronomy and lead the new development of major scientific instruments for astrophysics research. We intend to develop a small-scale instrument, optimize it, and then use the demonstrated performance as a centerpiece for several new directions, which could include a major instrument for ground-based astronomy.

For this project, we propose developing the capability to produce large CCD-mosaic cameras, from the design stage, to a complete scientific camera that can be used at a major telescope facility. The design would be applicable to many of the next-generation ground- and space-based observatories. The currently available CCD camera designs are not as sophisticated—in their mechanical, electronics, readout, or computing designs—as the state-of-the-art high energy physics detector systems. Thus, at Berkeley Lab, we are well suited to developing very-large-area CCD arrays that can be used for very-deep high-resolution surveys of the early universe. The development would be in the context of creating a large instrument for a world-class wide-field telescope. This would allow us to aggressively pursue the uniqueness of our technology while exploiting the strengths of Berkeley Lab in creating large multi-faceted instruments.

### Accomplishments

For large-scale cameras for astronomy applications, we proposed to proceed in three main steps:

- First, to develop the lab-bench test capabilities necessary to illuminate and read out a mosaic of CCDs and to tune their performance characteristics.
- Second, electronics and mechanical camera design of a 3 by 3 mosaic of large-format CCDs, the dewar, and the testing of component parts and design elements.
- Third, camera assembly, and testing of the full 3 by 3 mosaic of large-format CCDs.

The first step is now complete. We have built the necessary clean room facilities and have acquired liquid

nitrogen dewars needed to cool the CCDs. We have purchased two commercial CCD controllers and interfaced them to a Sun workstation for testing. We have enhanced the commercial software for the controllers to automate many of the measurements needed for full testing of the CCDs. All CCD testing so far has been with CCDs mounted directly to conventional printed circuit boards (PCBs) or with indirect PCB mounting with an aluminum nitrate substrate for better cooling and thermal match. In both cases, the CCD signal pads are wire-bonded to the PCB and the signals routed to convention connectors. We have also been building models of final production mounts that support backside illumination where a CCD is glued directly to a compatible metal. The metal serves as the CCD support, acts as an attachment mechanism to the cooling system, and holds a PCB with connectors to which the CCD signals are wire-bonded. This scheme supports four-sided, close packing of the CCDs so that large-area arrays can be assembled.

The focus of the third-year activity will be on the design of mechanical aspects of a camera composed of a 3 by 3 mosaic of large-format CCDs. The project will include the testing of component parts and design elements. We will also proceed to camera assembly, and testing of the full 3 by 3 mosaic of large-format CCDs.

---

### ***Sub-Project 00027: Nearby Supernova Search with Ten-Fold Increase in Efficiency***

---

#### **Project Description**

The study of cosmology has entered a new era in which empirical data can play a decisive role in determining the correct theory of the history and fate of the universe. While previously this theory was heavily dependent on esthetic considerations, we now are beginning to have a range of experimental /observational tools that can directly measure the relevant cosmological parameters. Among these tools, supernovae stand out as potentially the most direct, least model-dependent, for studying the energy-densities of the universe and the relative contributions of mass energy and vacuum (or "dark") energy. Over the past 15 years, Berkeley Lab has developed this supernova tool to the point that its current main result—the evidence for the existence of significant vacuum energy—is now considered to be a crucial element of the current State of the Universe report by much of the astrophysics community.

The next key step is to take the range of techniques that we have developed and perform the definitive measurements that will make the supernova results one of

the solid foundations on which future cosmological investigations will build. By studying an order-of-magnitude more supernovae in a much larger, systematic program, we can address each of the main remaining sources of uncertainty (primarily systematics). The "supernova values" for the mass density, vacuum energy density, and curvature of the universe will then become the benchmarks for the other, more model-dependent cosmological measurement techniques. In particular, the cosmic microwave background measurements will be able to use these values as both a starting point and a benchmark as they fit the eleven or so parameters to which their power spectrum is sensitive.

This project will provide Berkeley Lab with the capability to dramatically scale up discovery of nearby supernovae. Instead of finding dozens of supernovae during a semester, we would gear up the search to discover several hundred and therefore are calling this a Supernova Factory (SNfactory). We are learning how to do this in an efficient manner, and learning which telescopes are most suitable for the task. In particular, the NEAT [Near Earth Asteroid Tracking program operated by the Jet Propulsion Laboratory (JPL)] search runs are going to increase. Over a three-month span, we would expect NEAT to find over 200 supernovae in the Hubble Flow. In addition to NEAT, the LINEAR (Lincoln Near Earth Asteroid Research) team will be surveying the entire visible sky twice a month. Discussions concerning a collaboration are underway and we will look to characterize their telescope in the near future for use in a supernovae search.

We must greatly improve the computational tools for managing the dataflow, data analysis, scheduling, and multi-telescope coordination. We have identified the following items as critical tasks. They are all feasible, but will require innovative large-data-set handling, and novel software for coordination of an international collaboration of humans, computers, and telescopes.

- *More complete automation of the search.* Just controlling and monitoring the dataflow, required most of the time of two experienced scientist/programmers during the period of the pilot study search. This will have to be automated for year-round searching.
- *Automation of target sky-field selection, and distribution to the appropriate telescopes (e.g., NEAT).* We will need automation of the evaluation process of search observations and appropriate rescheduling.
- *Improve automated candidate pre-screening by at least a factor of 30.* We will not be able to repeat our current feat of 30,000 candidates searched by eye, and we need to be able to handle at least 10 times more data.

- *Develop more reliable network connections and use dedicated machines.* We will need automation of follow-up observation scheduling for the multiple telescopes that will observe the supernovae after discovery. Automation is also needed of analysis of follow-up data feedback of these results for the planning and scheduling of further follow-up observations.
- *Queue scheduled spectroscopy and photometry (VLT, Gemini North and South SOAR).* The final siting and operation of our own Berkeley Lab 30-inch telescope at Chews' Ridge, California, will provide a northern hemisphere automated queue-scheduled photometry telescope.

## Accomplishments

The project depends on the use of the Near Earth Asteroid Telescope (NEAT) to find large numbers of supernova candidates. Part of our effort has focused on developing improved supernova-searching capabilities with NEAT. The NEAT Team at JPL has moved to a larger telescope (1.2-meter diameter, formerly 1.0-meter diameter) on Haleakala, Hawaii, and has increased the number of search nights from 6 to 18 per month. The telescope is now equipped with a re-imaging camera built by Boeing for the United States Air Force. NEAT has also added water cooling (previously thermal-electric cooling) to its CCD detector, enhancing its sensitivity to faint targets by lowering the dark current. This facility is to be the primary source of supernova discoveries for the SNfactory, so these improvements lead directly to improvements in the baseline performance of the SNfactory. Besides the greater depth attainable with the improved NEAT, it will be possible to search patches of sky for supernovae more frequently, allowing supernovae to be discovered much earlier after explosion.

Since NEAT has resumed search operations with these new capabilities, we have provided them with scripts needed to automatically transfer imaging data from Haleakala to Berkeley Lab for processing, and these scripts have been tested on a run of real data. A draft Memorandum of Agreement has been developed and will be concluded soon. We will also use NEAT facilities at the Palomar Observatory and we have worked with Caltech to install the network infrastructure needed to support this use on the Palomar telescope. This currently uses a T1 land connection but we are exploring the use of high-speed wireless to provide higher throughput.

Another priority is the acquisition of a telescope for follow-up of nearby supernovae. We have completed negotiations with the University of Hawaii (UH) regarding the use of their 2.2-meter telescope on Mauna Kea in Hawaii to obtain follow-up observations (spectra and

lightcurves) for supernovae discovered by the SNfactory using NEAT data. A Memorandum of Agreement is now in place to ensure that we will have 20% access to the telescope. Mauna Kea is the world's premier site for astronomical observations, providing the best image quality available from the ground.

The major instrumentation effort has gone into creating baseline design for a supernovae follow-up spectrograph. The SNfactory concept for an integral field unit (IFU) spectrograph for use in obtaining supernovae spectra and lightcurves has been much more fully developed. With the help of the OASIS/Sauron group at the Observatory of Lyon (France), a prototype optical and mechanical design has been developed for an IFU spectrograph for the UH 2.2-meter telescope. This spectrograph will have both a blue and a red optical channel, allowing optical elements and detectors to be optimized for each channel. The estimated "average" efficiency for this spectrograph of 20% (including telescope and atmosphere) is better than the typical "peak" efficiency of spectrographs currently available. Because the integral field unit obtains a spectrum of the 122 0.5 by 0.5 sections of sky from the 6 by 6 arcsecond region of sky around and including each supernova, all the supernova light is captured. This allows not only spectroscopy, but also synthetic photometry, to be obtained. Synthetic photometry is obtained over the entire optical spectral region, making this approach much more efficient than typical photometry, which involves serial observations in up to six separate filters. Because, for synthetic photometry, filter bandpasses can be defined in software, the photometry can be more precise and can be checked for a wider range of potential systematic errors. Moreover, the large region covered by the IFU means that precise pointing of the telescope is not required. Therefore, we expect to be able to automate the acquisition and subsequent observation of each supernova. This minimizes our reliance on a night assistant at the telescope; the night assistant will need to do nothing more than point the telescope using standard procedures (accurate to typically a few arcseconds) and then press a mouse button to commence observations. This instrument will be truly revolutionary. Funds and manpower in France have been earmarked for construction of the first follow-up spectrograph. A Memorandum of Understanding between Lyon and Berkeley Lab has been drafted and will be concluded soon.

A key feature of the project is new supernova search software. Graduate student Michael Wood-Vasey has implemented an adaptive kernel image subtraction package into the code used to discover supernovae in the NEAT images. This is a critically important addition since the wide field covered by the NEAT images can result in changes in the point-spread function (PSF) over the field. Images taken weeks apart can have different PSF gradients, leading to subtraction errors and the detection of spurious supernova candidates. Since, on average, there

will be only one supernova per 25 NEAT images, it is critical that the number of spurious candidates be kept to much less than one per NEAT image. Spurious candidates can usually be eliminated by human inspection, but the flow of data from NEAT will be so large that the commitment of manpower would be ridiculous without this important control. This improved software is now being tested on real NEAT data.

---

## **Large Astrophysical Data Sets**

---

Principal Investigators: George Smoot and Jodi Lamoureux

Project No.: 99040

### **Project Description**

In the past decade astrophysics has undergone a renaissance, transforming from a data-starved science to a data-driven one. The advent of new technologies including satellites and large earth-based detectors are generating massive data sets. The Cosmic Background Explorer COBE satellite and subsequent balloon-based observations of the Cosmic Microwave Background have begun to give us a detailed picture of the universe at its earliest moments; the Hubble Space Telescope and the Keck Telescope have found galaxies at distances corresponding to the universe at one-tenth of its present age; the Two Degree Field (2DF), Center for Astrophysics (CfA) and APM large-scale red-shift surveys have begun to map out the present structure of the nearby universe. Thanks to the large investments that agencies such as National Science Foundation (NSF), National Aeronautic and Space Administration (NASA), Department of Energy (DOE), and European Space Agency (ESA) are making in astrophysics and cosmology, the amount of data on all fronts will increase by orders of magnitude in the coming decade, enabling us to determine the history of the universe and understand its fascinating constituents with unprecedented precision.

However, the size and complexity of these data sets threatens to leave the community data-swamped. For example, a typical current astronomical charge-coupled device (CCD) camera, 4096 by 4096 pixels, digitized to 18 bits, produces  $3 \times 10^8$  bits per picture and often takes enough pictures to produce several gigabytes of data per night. Other large-area and -volume detectors easily produce data at that rate. For example, a neutrino detector

with 5000 phototubes triggering at a kilohertz will produce about 30 gigabytes of data each day.

Existing algorithms and hardware will simply be overwhelmed by the sheer volume of the new data. Typical reconstruction algorithms currently available process three to four events per second. The realization of our scientific goals will depend crucially on meeting the qualitatively new computational challenges set by the quantitatively new data. The issues we face—in the analysis, synthesis, and presentation of the data, data compression and transmission, mass storage, data mining, parallel algorithms, scaling ability, and complex data visualization—are also at the forefront of current research in computer science.

A group of us on the University of California, Berkeley campus and at the National Energy Research Scientific Computing Center (NERSC) are developing a focused collaboration among astrophysicists to address the computational tools, techniques, and technologies needed to cope with the new challenges posed by future astrophysical data sets. We expect that our needs as astrophysicists will provide a practical spur to new developments in computer science, in turn enabling new research both within astrophysics and more widely in other data-intensive disciplines.

### **Accomplishments**

We have focused on a number of large astrophysical data sets for experiments like the Antarctic Muon and Neutrino Detector Array (AMANDA), IceCube, Supernova Factory, and SuperNova/Acceleration Probe (SNAP). AMANDA is the current operational neutrino telescope with ~8 terabytes of data from 1996 to 2001 stored in High Performance Storage System (HPSS). IceCube is in the proposal stage, and we have designed the Data Handling Software for it. We have worked with the Supernova Factory project to understand the data model and currently available software. Based on this, we have made careful extrapolations to define the proposed SNAP workflow.

### **Software Tool Development**

In previous years, we built a number of Data Handling Tools that we applied to the AMANDA data sets. These include an automation library (perl) to organize production processing, a server/client Java data distribution applet that downloads files from HPSS to our international collaboration, and a new three-dimensional visualization program based on OpenGL. Screen shots of AMANDA and IceCube events may be viewed at: <http://rust.lbl.gov/~jodi/iceCube/vis.html>.

### AMANDA Data Mining

Since 1998, we have processed the 1997 AMANDA data sample twice, the 1998 data sample once, and we are currently finishing up the production processing of the 1999 data sample. Data from all years can be downloaded from our distribution servers. As AMANDA ramps down and IceCube ramps up, we expect to streamline the data processing and distribution system so that these tasks are done in real time.

### Scientific Calculations

Several scientific calculations have been important to this Laboratory Directed Research and Development (LDRD) project. We have submitted a paper entitled, "Astrophysical Neutrino Event Rates and Sensitivity for Neutrino Telescopes" for publication. This can be used as a guide for predicting data rates in all currently existing and planned water or ice Cherenkov detectors. The paper

is scientifically interesting because it calculates the level of neutrino fluxes, which can be discovered presently and in the foreseeable future. In addition, we have begun estimating the signal-to-background fraction for the SNAP trigger. Efficient operation of the SNAP mission depends on having very low contamination of SuperNova (SN)-1b, SN-1c, and SN-II in the SN-1a sample. The data flow and complexity of the experiment control are very sensitive to this fraction. It is also strongly correlated to the precision with which SNAP can determine the fundamental cosmological parameters which makes it scientifically interesting.

### Design Work

This year we also focused on designing the Data Handling/Mining systems for future telescopes. Both IceCube and SNAP are good candidates for funding and both depend strongly on NERSC for software development support and high-performance computing.

## Up-going muon from 1997 AMANDA data, South Pole

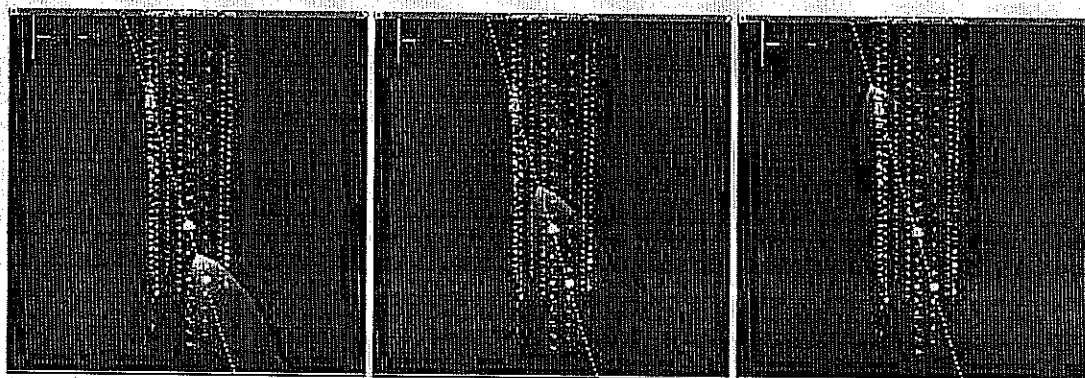


Figure 40: Three views at successively later times showing the Cherenkov light cone from a muon passing through the AMANDA detector in 1997. This muon has come through the earth and is exiting the South Pole Icecap with a 30-degree angle from the zenith. The event moves from lower right toward upper left. Each white ball shown is an optical sensor in the AMANDA detector..

### Publications

I. Albuquerque, J. Lamoureaux, and G. F. Smoot, "Astrophysical Neutrino Event Rates and Sensitivity for Neutrino Telescopes" (August 2001), submitted to *Astrophysical Journal*.





## Acronyms and Abbreviations

ADC	analog to digital converter	EPS	electronic phase separation
AFM	atomic force microscopy	ES	embryonic stem
AFRD	Accelerator and Fusion Research Division	ESA	European Space Agency
ALS	Advanced Light Source	ESD	Earth Sciences Division
AMANDA	Antarctic Muon and Neutrino Detector Array	ESDG	Earth Science Data Grid
AMR	Adaptive mesh refinement	EXAFS	extended x-ray absorption fine structure
APC	antigen presenting cell	FHWM	full width at half maximum
ARPES	angle resolved photoemission spectroscopy	FICM	fluorescence interference contrast microscopy
BGS	Berkeley Gas-filled Separator	fnRNA	functional noncoding RNA
BNCT	Boron Neutron Capture Therapy	FRET	fluorescence-resonance-energy transfer
BOOMERANG	Balloon Observations of Millimetric Extragalactic Radiation and Geophysics	fs	femtosecond
CCD	charge-coupled device	FTIR	Fourier transform infrared spectroscopy
CCSM	Community Climate System Model	FY	fiscal year
CERN	the European Laboratory for Particle Physics near Geneva, Switzerland	GFP	green fluorescent protein
CH	Chemical Sciences Division	GGA	generalized gradient approximation
CHD	coronary heart disease	GRETA	Gamma-Ray Energy Tracking Array
CMB	cosmic microwave background	GUPS	giga-updates-per-second
CMR	colossal magnetoresistive	HEP	High Energy Physics
CPU	central processing unit	HPSS	high performance storage system
CQW	coupled quantum well	HRM	hierarchical resource manager
CS	Computing Sciences	HTSC	high temperature superconductivity
cw	continuous wave	IR	infrared
DFT	density functional theory	IRAM	intelligent RAM
DNA	deoxyribonucleic acid	ISA	instruction set architecture
DOE	Department of Energy	JGI	Joint Genome Institute
DPS	Diesel Particle Scatterometer	JPL	Jet Propulsion Laboratory
DRM	disk resource manager	LBNL	Ernest Orlando Lawrence Berkeley National Laboratory
DSB	double strand break	LDA	local density approximation
DSP	digital signal processor	LDOS	local-density-of-state
ED	Engineering Division	LDRD	Laboratory Directed Research and Development
EDTA	ethylenediaminetetraacetic acid	LF	light fraction
EETD	Environmental Energy Technologies Division	LFU	least frequently used
EM	electromagnetic	LHC	Large Hadron Collider, CERN

l'OASIS	laser Optics and Acceleration Systems Integrated Studies	psi	pounds per square inch
LP	lipoprotein	PVP	parallel vector processor
LRU	least recently used	rf	radio frequency
LSD	Life Sciences Division	RHEED	reflection high energy electron diffraction
LSI	latent semantic indexing	RHIC	Relativistic Heavy Ion Collider, Brookhaven National Laboratory
LWFA	laser wakefield accelerator	RNA	ribonucleic acid
MAXIMA	Millimeter Anisotropy eXperiment Imaging Array	RTC	Recoil product Transfer Chamber
MCF	Macromolecular Crystallography Facility, ALS	RXFH	Resonant-XFH
MEFS	mouse embryonic fibroblasts	SC	Office of Science, DOE
MES	molecular environmental science	SC	superconductivity
MHC	major histocompatibility complexes	SEM	scanning electron microscope
MSD	Materials Sciences Division	SLAC	Stanford Linear Accelerator Center
NASA	National Aeronautics and Space Administration	SLIM	structured light-imaging microscope
NCAR	National Center for Atmospheric Research	SNAP	SuperNova/Acceleration Probe
NERSC	National Energy Research Scientific Computing Center	SOI	silicon-on-insulator
NEXAFS	near-edge x-ray adsorption fine structure	SOM	soil organic matter
NIH	National Institutes of Health	spFRET	single-pair FRET
NMR	nuclear magnetic resonance	SQUID	Superconducting QUantum Interference Device
NOAA	National Oceanic Atmospheric Administration	ss-DNA	single stranded DNA
NSD	Nuclear Science Division	STAR	Solenoidal Tracker at RHIC
NSF	National Science Foundation	STM	scanning tunneling microscope or microscopy
OM	organic matter	SVD	singular value decomposition
PBD	Physical Biosciences Division	SXH	soft x-ray holography
PCA	principal component analysis	TCR	T-cell receptors
PCB	printed circuit boards	TES	transition-edge sensors
PCR	polymerase chain reaction	TRLFS	time-resolved laser fluorescence spectroscopy
PD	Physics Division	tRNA	transfer RNA
PDB	Protein Data Bank	UC	University of California
PET	positron emission tomography	UHV	ultra-high vacuum
PI	principal investigator	UV	ultraviolet
PIM	processor-in-memory	VLDL	very low density lipoprotein
PIN	positive-intrinsic-negative	VOC	volatile organic compound
POLARBEAR	POLARization of the Background millimEter background Radiation	VSOM	visual servoing optical microscopy
PSF	point-spread function	XANES	x-ray absorption near-edge structure spectroscopy
		XFH	x-ray fluorescence holography